Bias correction methods are variations on downscaling that have been developed to address spatiotemporal bias in the CMAQ model. For example, Crooks and Oezkaynak (2014) developed a statistical method of spatiotemporal bias correction of $PM_{2.5}$ mass and its major components for CMAQ fields. The correction uses speciated data from ambient monitors. Mass conservation for $PM_{2.5}$ observations constrains the sum of the $PM_{2.5}$ components' concentrations in locations without speciation monitors. The Crooks and Oezkaynak (2014) method is similar to downscaling methods in that it is a calibration method, but it corrects to the grid-scale rather than receptor points. The method was developed for use in an epidemiologic study investigating the association between $PM_{2.5}$ component ambient concentrations and birth outcomes throughout the state of New Jersey based on 1-month averages, so the focus was on addressing seasonal bias trends rather than daily biases. The bias-corrected CMAQ results were more accurate than the original CMAQ output (calculated as mean bias and RMSE using monitored concentrations as a reference), and a cross-validation study found that predictions improved when enforcing mass conservation. Comparison between the bias-corrected CMAQ and other downscaling or bias correction methods was not provided. Hogrefe et al. (2009) used a combined model-observation approach to estimate historic gridded fields of $PM_{2.5}$ mass and component concentrations, with corrections varying by component, season, and location. $PM_{2.5}$ mass concentration had a median bias of $-0.3$ $\mu g/m^3$ and median RMSE of 7.5 $\mu g/m^3$ compared with monitor values. Hogrefe et al. (2009) reported high relative biases and larger uncertainties for nitrate and organic carbon, compared with sulfate and ammonium. This was especially pronounced at remote IMPROVE sites, compared with urban CSN sites that have more monitors. Although more development is needed, these methods present additional options for applying CTMs for modeling $PM_{2.5}$ species.

A hierarchical Bayesian model (HBM) to predict daily $PM_{2.5}$ exposure concentrations for use in the Environmental Public Health Tracking Network has been developed through a CDC-EPA collaboration. This model integrates U.S. EPA monitor data with CMAQ simulation results to generate daily $PM_{2.5}$ concentration and error fields for a 36 km grid across the conterminous U.S. and for a 12 km grid across an eastern portion of the U.S. (Vaidyanathan et al., 2013; McMillan et al., 2010). In the application of HBM over a section of the eastern U.S., McMillan et al. (2010) found that the mean squared error using the HBM field was similar to a field developed using kriging, though the HBM outperformed kriging by 10–15% for bias. They found that 59% of the validation data was captured in the kriging prediction intervals as compared to 80–90% when using HBM. For the U.S.-wide application at 36 km resolution, the HBM method had Pearson $R$'s ranging from 0.91 to 0.94, depending upon the method used to impute the CMAQ data (Vaidyanathan et al., 2013), while the 12 km application over the eastern portion had Pearson $R$'s of 0.84 to 0.86.

Data fusion methods sometimes include fusing CTM modeling results with observations for exposure predictions. Chen et al. (2014) evaluated an observation-CMAQ fusion for population air pollution exposure assessment using an inverse distance weighting method on observation-CMAQ differences, concluding that data fusion improved the estimation of population-weighted average exposure concentrations. On average, $PM_{2.5}$ mass was estimated to be negatively biased by about 30%,

and individual components had a range of positive and negative biases from −150 to 100%. Nitrate and OC tended to see the largest biases and errors. After data fusion, the bias for $PM_{2.5}$ was near zero. Performance for individual components was similarly improved. Friberg et al. (2016) also fused CMAQ results to observations in a study focused on $PM_{2.5}$ exposures in Georgia. In this study, daily spatial exposure concentration fields for $PM_{2.5}$ mass, $PM_{2.5}$ components, and various gases were constructed from two blended fields. For one field, the temporal variance is driven by observations, while the spatial structure is driven by the annual mean CMAQ fields. The second field is constructed by scaling daily CMAQ simulated fields using mean observations to reduce bias. The final step blends the two fields based on using the temporal variance. The method intentionally does not force the fields to the observations at each monitor as they can be impacted by local emissions. The original CMAQ application for $PM_{2.5}$ was biased low about 12% with an RMSE of about 50% and an $R^2$ of 0.3. Typically, performance for individual $PM_{2.5}$ components was not as good. After applying the data fusion, the bias was almost totally removed, the RMSEs were about 20% for $PM_{2.5}$ and most PM components (though $NO_3^-$ and EC were substantially higher), and the $R^2$ was about 0.92 (similar to individual components, though $R^2$ for EC was about 0.8). The method was tested using a 10−fold cross validation. In this case, the $PM_{2.5}$ $R^2$ was 0.75 and the RMSE was 30%.

Data fusion techniques have been tested in several other locations. Friberg et al. (2017) compared the fused CMAQ with original CMAQ model runs for five cities (Atlanta, GA, Birmingham, AL, Dallas, TX, Pittsburgh, PA, and St. Louis, MO) and found that the RMSE for $PM_{2.5}$ ranged from 2.21 to 3.76 $\mu g/m^3$ for the fused CMAQ, compared with 6.93 to 7.86 $\mu g/m^3$ for the original CMAQ.Huang et al. (2018) applied this method to North Carolina. In addition to doing the traditional 10-fold cross-validation, they also used spatial grouping of the 10% of monitors being removed to account for monitor clustering. In this case, the simulated $PM_{2.5}$ from the base CMAQ application had an RMSE of 6.3 $\mu g/m^3$ and an $R^2$ of 0.3, while after data fusion the RMSE decreased to 1.8 $\mu g/m^3$ and $R^2$ improved to 0.95. They also conducted 10-fold cross validation, both with and without (i.e., randomly withheld) spatial grouping. Finally, they compared the CMAQ-based data fusion fields with fields developed using a Bayesian-based method incorporating aerosol optical depth (AOD) from satellite data and found that the CMAQ-based approach performed slightly better (e.g., $R^2$ of 0.97 vs. 0.90 for AOD) using all of the data. The application of the same method in multiple locations shows that performance varies by domain.

Hybrid approaches can involve merging CTMs with dispersion and/or LUR models, merging CTMs with observational data, or some combination therein. Hybrid approaches improved CTM validation for $PM_{2.5}$ mass concentration when CTM was merged with either models or observational data. However, validation was not as good for $PM_{2.5}$ mass components, possibly due to the sparseness of validation data and limited data for $PM_{2.5}$ component emissions.

### 3.3.3 Satellite-based Methods for Exposure Concentration Estimation

At present, spatiotemporal methods for predicting exposure concentration based on satellite observations have been applied primarily to $PM_{2.5}$ using AOD information supplied by various satellite-based instruments [see Section 2.4.4 and (Lin et al., 2015; Hu et al., 2014c; van Donkelaar et al., 2014; Lee et al., 2012a; Mao et al., 2012; Liu et al., 2009)]. Satellite data (Section 2.4.5), obtained twice per day over the U.S., has been used in recent exposure assessment studies to estimate exposure concentrations in rural regions where monitoring is not conducted, to improve estimates of spatial variability in exposure concentrations, and to cover larger geographic regions. For example, Hystad et al. (2012) used a composite satellite image of AOD over the years 2001 to 2006 to estimate $PM_{2.5}$ exposure concentration across Canada, which includes urban and rural areas. The authors adjusted the satellite data by annual average $PM_{2.5}$ (or estimated $PM_{2.5}$ based on TSP measurements prior to $PM_{2.5}$ measurements, which began in 1984) and then used the study cohorts' residential locations to estimate their exposures based on their residential histories and exposure concentrations corresponding to those locations. Hystad et al. (2012) noted that incorrect assignment of exposure based on failure to account for movement between residences over time and space through this method resulted in 50% of individuals being classified in the wrong $PM_{2.5}$ exposure quintile. Prud'homme et al. (2013) computed the correlation of $PM_{2.5}$ exposure concentration predicted at a residential location with the nearest fixed-site monitor and found that the correlation decreased from $R = 0.74$ (not stated if Pearson or Spearman) when the home was within 1 km of the monitor and decreased to 0.60 for distances of 30−40 km between the home and the monitor. This result implies that the $PM_{2.5}$ exposure concentration predicted using AOD is a better predictor of exposure concentration within a given grid cell compared with exposure concentrations further away.

Errors in the relationship between $PM_{2.5}$ and AOD are related to variation in retrieval due to resolution of the satellite image and variation in meteorology, topography, and reflectance (Section 2.4.4). Hu (2009) calculated the correlation between surface $PM_{2.5}$ and AOD at 877 monitoring sites across the U.S. and found that average correlation east of the 100°W longitude line was Pearson $R = 0.67$, compared with Pearson $R = 0.22$ west of the 100°W longitude line. Negative correlations between $PM_{2.5}$ and AOD were calculated at several sites west of the 100°W longitude line but at only three locations east of the 100°W longitude line. van Donkelaar et al. (2010) also noted this discrepancy between satellite data quality in the eastern and western U.S. They used population-weighting to determine national and global estimates of exposure concentration. Population density happens to be lower in mountainous parts of the western U.S., where the highest biases in AOD were noted.

Improving the relationship between AOD and surface PM observations to estimate exposure concentrations has led to the use of more advanced statistical methods for fusion of satellite data with CTM output and surface data in recent years. Satellite-based exposure concentration models now use AOD and other information (e.g., direct pollutant observations, meteorology, and land-use). For example, van Donkelaar et al. (2012) applied a smoothed bias correction to satellite-derived $PM_{2.5}$ exposure

concentrations by first applying a 90-day moving average to the AOD prior to fitting $PM_{2.5}$ concentration estimates, and then smoothing the $PM_{2.5}$ exposure concentration field using IDW. The bias correction alone reduced the positive bias in the estimate to +29% with an estimated uncertainty of 54%. This is compared to the uncorrected $PM_{2.5}$ exposure concentration estimate, which had a bias of 97% with an estimated uncertainty of 67%. Incorporation of smoothing reduced the bias further to +14% with an uncertainty of 42%. An LUR approach to derive spatiotemporal pollutant fields accounts for the complexities in the AOD-PM relationships, including spatially and temporally varying conditions (Lee et al., 2016; Hu et al., 2014e; Ma et al., 2014; Chudnovsky et al., 2012; Hystad et al., 2011). Similar to LUR models, the approach is to develop a regression relationship between the observed $PM_{2.5}$ and AOD that includes the AOD field available from satellite observations and, potentially, other variables (e.g., those used in traditional LUR modeling). The regression coefficients can vary in time and space.

Not accounting for spatial and temporal variability in the relationship between $PM_{2.5}$ and AOD may lead to poor model performance (Hu et al., 2014d). Liu et al. (2009) recommended use of a two-stage general additive model including land use variables, with a stage one temporal model and stage two spatial model, so that the temporal and spatial variability are both addressed by the model, with an out-of-sample $R^2$ of 0.78, which was close to the model fit $R^2$ of 0.79 (stage one model-fit $R^2 = 0.77$, stage two model-fit $R^2 = 0.73$). Given the large spatial and temporal coverage of satellites, a large number of observations are typically available to develop the model. Additional spatial variation, particularly at scales finer than the resolution of the satellite observations, is provided by using fine scale land use variables. Lee et al. (2011) also recognized that the relationship between $PM_{2.5}$ and AOD is governed by time varying parameters affecting the vertical profile, the temporal variability of surface $PM_{2.5}$ over the course of a day. They developed a day-specific mixed effects model with random intercepts and slopes to quantify the relationship between surface $PM_{2.5}$ measured by surface monitors and AOD over New England in 2003. They assumed that temporal variability in properties that most strongly affect this relationship are much larger than their spatial variability over the domain of interest. In their model, the AOD fixed effect represents the average effect of AOD on $PM_{2.5}$ for all study days and the AOD random effects explain the daily variability in the $PM_{2.5}$-AOD relationship. Since some ground-based $PM_{2.5}$ monitors are located near strong sources, but Moderate Resolution Imaging Spectroradiometer (MODIS) samples represent an average over a 10 km × 10 km grid, an additional site specific random effects term is added to correct possible bias. Site specific out-of-sample $R^2$ varied from 0.87 to 1.0 with precision ranging from 8.8 to 38.6% for measured mean $PM_{2.5}$ at 26 urban sites (range: 9 to 19.5 $\mu g/m^3$).

Satellite observations of AOD have also been incorporated into hybrid modeling approaches. For example, Beckerman et al. (2013b) combined LUR, based on AOD observations, GEOS-Chem model output, land use data, and surface measurements of $PM_{2.5}$ concentration, with BME to predict $PM_{2.5}$ concentrations. BME was added to the model to improve spatiotemporal variability at scales smaller than the satellite's spatial resolution. Beckerman et al. (2013b) did not observe a substantial added benefit to including satellite data in an LUR model that also drew from land use data, surface measurements of $PM_{2.5}$ concentrations, and GEOS-Chem simulations. In this study, $PM_{2.5}$ concentrations were predicted

throughout the contiguous U.S. using an LUR-BME with and without satellite data. The LUR with inclusion of satellite data produced an out-of-sample $R^2$ of 0.27 compared with $R^2$ of 0.05 without inclusion of satellite data. When BME was incorporated in the LUR to interpolate between spatiotemporal residuals from the training model, out-of-sample $R^2$ improved to 0.79. $R^2$ was the same for the simulations both including and excluding satellite data. Using a similar hybrid satellite-modeling approach, Lee et al. (2012a) found that during the period 2000–2008 in the New England region of the U.S., a densely populated study domain with high traffic areas, $PM_{2.5}$ exposure concentrations were predicted with an out-of-sample $R^2$ value of 0.83 and a mean relative error of 3.5%. Chang et al. (2014) describe a statistical downscaling approach that incorporates LUR models utilizing AOD and statistical techniques for combining air quality data sets that have different spatial resolutions. In cross-validation experiments for a 3-year time period over the southeastern U.S., the model performed well (out-of-sample $R^2 = 0.78$ and RMSE = 3.61 $\mu g/m^3$ between observed and predicted daily $PM_{2.5}$ concentrations), with a 10% decrease in RMSE attributed to the use of AOD as a predictor. Validation of hybrid models has been inconsistent across studies.

Recent studies have tested the effect of satellite image resolution on $PM_{2.5}$ mass concentration predictions. Hu et al. (2014c), using a two-stage model, compared the more traditional MODIS AOD at 10 km resolution with a Multiangle Implementation of Atmospheric Correction (MAIAC) algorithm at 1 km in the Southeastern U.S. and found that, when using 10-fold cross-validation, the out-of-sample $R^2$ was slightly lower for the 1 km MAIAC observations (0.67 vs. 0.69), though the $R^2$ for model fitting was the same (0.83). This can be contrasted against Chudnovsky et al. (2013), discussed in Section 2.4.4 Alexeeff et al. (2015) also used the 1 km MAIAC fields to estimate exposure concentration fields, comparing their results to fields developed using kriging. They found that using the MAIAC-based fields had a higher cross-validation than kriging, and that the low out-of-sample $R^2$ yielded biases in areas with lower covariance in the concentration field. Lv et al. (2016) used MODIS AOD and a statistical method similar to Chang et al. (2014) in an application in China. It is discussed here in terms of how the evaluation was performed. Using all data (no withholding), the $R^2$ was 0.78 and the normalized mean error was 0.27. When they used a random leave 10% out procedure, the method led to an $R^2$, normalized mean error (NME) and RMSE of 0.68, 0.26 and 21.40 $\mu g/m^3$, respectively (like $PM_{2.5}$ concentrations, RMSE is much higher in China than in the U.S.). Using a process where monitors were removed after being grouped by city led to somewhat worse performance: 0.61, 0.28 and 23.53 $\mu g/m^3$, respectively. This suggests that method and application evaluations should use cross-validation methods that consider spatial groupings of monitors as a more stringent evaluation approach.

Recent efforts have fused satellite data with LUR model results and surface observations to maximize available data for estimation of exposure concentrations. Kloog et al. (2011) built a three-stage regression model using surface measurements as the response variable and including MODIS-derived AOD, land use variables, and a daily calibration $PM_{2.5}$ concentration from surface measurements to estimate $PM_{2.5}$ exposure concentration on a 1 km × 1 km grid across New England, and Kloog et al. (2012a) extended the model across the Mid-Atlantic states. When AOD was available, the

cross-validation out-of-sample $R^2$ was 0.83 for New England and 0.87 for the Mid-Atlantic states; when AOD was unavailable, cross-validation out-of-sample $R^2$ was still 0.81 for New England and 0.85 for the Mid-Atlantic states. When running the model for the two regions combined, Kloog et al. (2012b) found cross-validation out-of-sample $R^2$ was 0.81 for the total model of $PM_{2.5}$ and 0.81 for the LUR stage of the model. Kloog et al. (2014) built upon this method by first calibrating the AOD on daily measurements of $PM_{2.5}$ and adjusting for land use and meteorological variables for the Northeastern U.S. (New Jersey to Maine) for 2003–2011. Where AOD data were available, this model was used to predict $PM_{2.5}$ exposure concentration. The second model used the AOD–$PM_{2.5}$ calibration to predict AOD, which was then input into the regression model for a 1 km × 1 km grid. Finally, a 200 m × 200 m resolution prediction was developed by taking the residuals at each monitoring site and regressing them against the fine-scale resolution predictors to estimate fine-scale $PM_{2.5}$ exposure concentration. The models were built separately for temporal and spatial variables, and each had an average cross-validation out-of-sample $R^2 = 0.87$.

Similar to BME, machine learning approaches can be used to merge satellite observations with land use and other data for prediction of $PM_{2.5}$ mass concentration. For example, Reid et al. (2015) used a machine learning approach to estimate spatiotemporal $PM_{2.5}$ exposure concentration fields over the central region of California during a period of wildfires in the region by building spatiotemporal models using 11 model types from a set of 29 independent variables and selecting the optimal one for each model type. Input data included $PM_{2.5}$ and meteorological predictions from a CTM (WRF-Chem), land use data, and satellite AOD observations [three sets: the Geostationary Operational Environmental Satellite West Aerosol/Smoke Product (GASP) with a resolution of 4 km, the MODIS AOD product with a resolution of 10 km, and a local AOD product developed from MODIS data at a 500 m resolution, $PM_{2.5}$ and meteorological predictions from WRF-Chem, land use data, and distance to the nearest fire cluster]. The data were put in to each of the methods to develop a best model. Ten-fold cross-validation out-of-sample $R^2$ ranged from 0.387 to 0.803, and RMSE ranged from 1.49 μg/m$^3$ to 2.03 μg/m$^3$. It was found that similar model performance (within 1.5% of the RMSE) was achieved using only 13 variables, compared with a model of all 29 variables, with highest out-of-sample $R^2$ and lowest RMSE. They found that the variable most correlated with the $PM_{2.5}$ observations was the GASP followed by the distance to nearest active fire cluster, then the local AOD product and WRF-Chem $PM_{2.5}$ contributed equally. Di et al. (2016a) used a similar approach for a model of $PM_{2.5}$ exposure concentration across the contiguous U.S. GEOS-Chem simulation results were merged with satellite data for AOD, surface reflectance, and aerosol absorbance index, as well as with surface data from monitors reporting to AQS and data for meteorology and land use. For 2000–2012, out-of-sample $R^2 = 0.84$ with RMSE of 2.94 μg/m$^3$. The relationship between predicted and measured $PM_{2.5}$ concentrations was approximately linear until measured $PM_{2.5}$ concentrations were above approximately 60 μg/m$^3$. At that point, the predictions were insensitive to measured $PM_{2.5}$, but limited $PM_{2.5}$ concentration data were available above concentrations of 60 μg/m$^3$. These studies illustrate that the most important variables change, depending on the scenario modeled and the specific variables included.

Several other studies have devised novel methods to fuse observational data and results from models for estimation of exposure concentrations. Pirani et al. (2014) performed Bayesian spatiotemporal modeling for the assessment of short-term exposure to $PM_{10}$ in London, U.K. using mass concentration measurements and output from the high spatial resolution air dispersion modeling system. They found exposure concentration estimates in urban areas are improved by including city-scale particle component and long-range transport component with covariates to account for residual spatiotemporal variation. Crooks and Isakov (2013) developed a novel method using wavelets to blend CMAQ, AERMOD, and observation fields to capture intra-urban transport of pollutants across a spectrum of spatial scales. They used it to estimate block group and zip code centroid exposure concentrations in Atlanta, GA and found that it captured the concentrations down to scales on the order of 100 m.

Several studies using AOD observations to predict $PM_{2.5}$ have been published in recent years. Progress in this approach includes incorporation of AOD with LUR, BME, and geostatistical modeling approaches that also may include surface measurements. Most applications of these hybrid models were designed to make comparisons across space for long-term exposure studies, where the temporal averages were more stable than for short-term exposure studies. Still, validation results across these studies were inconsistent, so attention must be given to the strengths and limitations of individual exposure models and their appropriateness for a given scenario (e.g., urban vs. rural, where monitoring for use in model training and validation may be sparse in the latter case) rather than assuming that the predicted $PM_{2.5}$ exposure concentration is accurate if it includes satellite data.

### 3.3.4 Microenvironmental Exposure Modeling

Indoor air exposures to total PM may be measured directly or estimated based on infiltration rates that typically use some level of mass balance model, potentially with chemistry, deposition, and other processes that can affect individual exposure. Inputs to indoor air mass balance models include ambient PM concentrations (observed or estimated), air exchange rates, indoor source emissions, and other factors that can affect the dynamics of pollutants. Such indoor air models are included in integrated exposure models (such as U.S. EPA's Stochastic Human Exposure and Dose Simulation [SHEDS] and Air Pollutants Exposure [APEX] models) or individual models (such as the Exposure Model for Individuals [EMI]), that also incorporate factors such as human activity patterns (Baxter et al., 2013). In Baxter et al. (2013), mean $PM_{2.5}$ exposure estimates obtained from models that considered time spent indoors and indoor-outdoor air exchange rates with no indoor sources were approximately half of the concentrations from ambient monitor measurements.

Personal exposure occurs in multiple microenvironments that people encounter through their daily activities (e.g., indoors, outdoors, in vehicles). Methods have been developed to simulate potential total exposures through such environments by tracking "representatives" of population groups as they move between indoor and outdoor microenvironments, using estimated pollutant concentrations in each

location to develop a time-weighted exposure profile for that population group. How individuals "move" though the different microenvironments is taken from studies of personal activity data [e.g., the Consolidated Human Activity Database, or CHAD (Isaacs, 2014)]. This database has information on sequential patterns of individual activities. This allows simulating not only "average" individual exposures, but also the distribution of exposures for different individuals or population groups over time.

Residential air exchange rate (AER) is a critical parameter for exposure models, such as APEX, SHEDS, and EMI (Breen et al., 2015; U.S. EPA, 2011, 2009a; Burke et al., 2001), with people spending the majority of their time indoors (Section 3.4.2.1). Since the appropriate AER measurements may not be available for exposure models, mechanistic, and empirical (i.e., regression-based) AER models can be used for exposure assessments. Empirical AER models do not consider the driving forces from the wind and indoor-outdoor temperature differences. Instead, a scaling constant can be used based on factors such as building age and floor area (Chan et al., 2005). Single-zone mechanistic models, such as the Lawrence Berkeley Laboratory (LBL) model, represent a building as a single well-mixed volume(Breen et al., 2010; Sherman and McWilliams, 2007; Sherman and Grimsrud, 1980). Recently, the LBL air infiltration model was linked with a leakage area model using population-level census and residential survey data (Sherman and McWilliams, 2007) and individual-level questionnaire data (Breen et al., 2010). Variations on the LBL model were compared with daily AER measurements in North Carolina (Breen et al., 2010) to find mean absolute differences of 40−43%.

The Hazardous Air Pollutant Exposure Model (HAPEM, now Version 6) is a screening level approach for modeling long-term inhalation exposures to ambient air pollutants, including PM. It can take modeled ambient pollutant concentrations as inputs or can use a parameterization of National Air Toxics Assessment (NATA)-generated PM estimates based on the near-road and far-from-road census tract populations (Rosenbaum and Huang, 2007). To develop exposure concentration estimates in microenvironments (e.g., commuting), microenvironmental factors are used to modify outdoor concentrations (e.g., provided by developing ambient exposure concentration fields). HAPEM has been used for nationwide assessments of exposure to sources of specific PM components and other pollutants (Ozkaynak et al., 2008) and, as noted above, coupled with a CMAQ/AERMOD combination (Isakov et al., 2009).

The SHEDS model and APEX model (which is now part of the Total Risk Integrated Methodology, or TRIM-Expo) both simulate individual movements though multiple microenvironments. APEX uses either a mass balance approach or a ratio to estimate in-vehicle or indoor concentrations (Che et al., 2015). Differences in subpopulation sampling methods between APEX and SHEDS produce small differences in predictions for population exposure concentrations (12.2 vs. 12.9 $\mu g/m^3$, respectively). SHEDS includes an activity-dependent ventilation rate to estimate dose. SHEDS-PM (the PM version of SHEDS) has a linear relationship between ambient concentrations and in-vehicle concentrations as well as in offices, restaurants/bars, schools, and stores. When analyzing contributions to exposure based on application of SHEDS-PM with daily $PM_{2.5}$ from CMAQ, Jiao et al. (2012) found that spatial variability

of ambient concentrations within urban areas was not substantial, but inter-individual variability in estimated exposures was substantial. Daily estimates of the ratio of ambient exposure to ambient concentration differed by a factor of 4–5 across the simulated individuals. SHEDS uses time-activity data from the CHAD database. Jiao et al. (2012) noted that there were not sufficient data in the CHAD database to quantify how time-activity patterns varied as a function of sex, region, or season when limited to the three areas studied, although statistically significant differences in time spent indoors or time spent outdoors by sex, region, and season were seen for CHAD data aggregated across large geographic regions. Liu and Frey (2011) proposed a method to estimate in-vehicle $PM_{2.5}$ exposure concentrations that combines using ambient concentrations and a local incremental concentration that accounts for near road enhancements in lieu of assuming a linear relationship between $PM_{2.5}$ concentration measured at fixed-site monitors and exposure concentrations estimated on the road using the CALINE4 dispersion model. Liu and Frey (2011) found that in-vehicle exposures contribute 10–20% of average daily $PM_{2.5}$ exposures. Georgopoulos et al. (2009) linked SHEDS with an environmental risk model (MENTOR) to estimate exposures (and the related risks) for $PM_{2.5}$ in Philadelphia, using a CTM to provide the $PM_{2.5}$ field. For those individuals with the highest 5% of $PM_{2.5}$ exposures, the major microenvironment was indoors, and environmental tobacco smoke was the dominant source. Ozkaynak et al. (2009) evaluated the uncertainty inherent in the coupled model formulation and compared it with a "crude" estimation of uncertainty when the models are run separately and with CMAQ outputs being used for SHEDS inputs. Uncertainty for the crude method was 1.2–4.4 times higher than for the coupled formulation.

The EMI model simulates individual exposure to $PM_{2.5}$ as the aggregate of exposures in multiple microenvironments (Breen et al., 2015). The EMI uses a five-tier system to model individual exposures. AER is predicted in Tier 1 based on surveys and variations on the LBL model for each microenvironment. Infiltration factors are predicted in Tier 2, and those values are used to predict outdoor concentrations infiltrated indoors measured immediately outside each microenvironment and measured at fixed-site monitors in Tier 3. A weighted average of the infiltration factor over time spent in different microenvironments is produced for each individual in Tier 4, and then personal exposures to pollution from directly outside the microenvironment and from the fixed-site concentration measurement are computed in Tier 5 for each individual. Personal monitoring and time-activity surveys are necessary inputs for the EMI. The Tier 2–5 metrics were observed to have approximately 15–25% error (Breen et al., 2018; Breen et al., 2015).

The trade-off between computational accuracy and efficiency in exposure and risk models has received limited discussion in the exposure model literature. Chang et al. (2012) described a simulation process incorporating SHEDS exposure simulation into two risk models: an "exposure simulator" in which an exposure time series was simulated stochastically and then incorporated into an ensemble average risk, and a two-stage "Bayesian" approach in which the computed time series was used as a prior in an exposure model. Risk of mortality (CHAPTER 11) associated with short-term $PM_{2.5}$ exposure was estimated using the exposure simulator model, the Bayesian model, and fixed-site $PM_{2.5}$ concentration as

an exposure surrogate. Little difference was observed between the exposure simulator and Bayesian models, but the exposure simulator was less computationally intensive.

### 3.3.5 Exposure Assignment Methods in Epidemiologic Studies

Epidemiologic studies use a variety of methods to assign exposures or exposure concentrations to study participants. Study design, data availability, and research objectives are all important factors for epidemiologists when selecting an exposure or exposure concentration estimation method. Common methods for estimating exposure concentrations from monitoring data include using fixed-site ambient monitoring, averaging concentrations from multiple monitors, and selecting the closest monitor to represent population exposure concentration. Investigators may also use statistical adjustment methods, such as trimming extreme values, to prepare the exposure concentration data set. Alternatively, modeling approaches described in Section 3.2.2 (modeling) can be used to estimate more spatially or temporally resolved exposure concentrations when data and resources are available.

Comparison studies have illustrated differences among the methods for producing estimates of exposure concentrations. For example, Dionisio et al. (2013) simulated $PM_{2.5}$ mass concentration, $PM_{2.5-EC}$, and $PM_{2.5}$–$SO_4^{2-}$ exposures or exposure concentrations using different methods including a fixed-site monitor, an AERMOD model, a hybrid model combining regional background estimates with local contributions by AERMOD, and the SHEDS exposure model. The methods differed more with respect to modeling spatial variability (as measured by coefficient of variation) compared with temporal variability, with spatial variability being greater for the AERMOD and hybrid approaches for all three pollutants. Temporal variability was similar across methods for $PM_{2.5}$ and $SO_4^{2-}$ with some difference across methods for EC. Mannshardt et al. (2013) compared use of fixed-site monitor concentration data, exposure concentrations estimated by CMAQ output, and exposures calculated using SHEDS to study respiratory emergency department visits associated with $PM_{2.5}$ exposure in New York County, NY, Queens, NY, and Bronx, NY. They found that the use of the SHEDS model led to a very similar relative risk as using CMAQ but provided additional information that helped reduce uncertainty. The effect estimates associated with exposure modeled by SHEDS and exposure concentration modeled by CMAQ were both higher and more precise than the effect estimate obtained from using fixed-site data as an estimate for exposure concentration. However, Mcguinn et al. (2017) estimated $PM_{2.5}$ exposure concentration and risks of coronary artery disease and myocardial infarction using a fixed-site monitor, CMAQ run with a census tract-level downscaler and with data fusion at 12 km resolution, and a satellite at 1 km and 10 km resolution. They did not find a relationship of model resolution with exposure concentration or with the magnitude of the effect estimates or with precision of the effect estimate for either health outcome studied.

Additional studies have also explored the effect of using different spatial averaging techniques to handle exposure concentration estimates from fixed-site monitoring data. Goldman et al. (2012) and

Strickland et al. (2013) compared exposure concentration estimates for $PM_{2.5}$, $PM_{10}$, $SO_4^{2-}$, $NO_3^-$, $NH_4^+$, EC, and OC among different methods, including fixed-site monitors, population-weighted averages of the (1) fixed-site monitors, (2) unweighted averages, (3) population-weighted averages, (4) area averages, and (5) a spatiotemporal model that used the pollutants' spatial and temporal autocorrelation structures to estimate exposure concentrations. Taking the spatiotemporal model as a reference, Goldman et al. (2012) found the fixed-site monitor had greater bias in the exposure metric compared with the averaging methods, and that bias increased for more-spatially-variable EC and OC compared with $PM_{2.5}$. These comparisons highlight differences among the methods in their ability to capture variability of exposures or exposure concentrations among study participants. The importance of capturing such variability also depends on the variability of the PM size cut or components.

Comparison of exposure concentration surfaces involving satellite observations have focused on spatial resolutions appropriate for different exposure concentration estimation techniques. Lee et al. (2012b) compared the appropriate averaging distance ranges for $PM_{2.5}$ exposure concentration surfaces estimated using satellite detection and kriging with $PM_{2.5}$ concentration measurements from fixed-site monitors using 6 years of data. Lee et al. (2012b) compared the kriged or remotely sensed data with the surface measurements over distances ranging from 7.6 km to 106.0 km using mean squared error (MSE), mean error, mean absolute error (MAE), Pearson correlation, and Spearman correlation. Lee et al. (2012b) estimated that kriging provided superior exposure concentration estimates when distances from the kriged estimate to the fixed-site monitor were smaller than 98 km while satellite detection provided superior exposure concentration estimates when distances from the remotely-sensed concentration centroid to the fixed-site monitor exceeded 98 km. Jerrett et al. (2016) compared remotely sensed $PM_{2.5}$ exposure concentration surfaces estimated from input by three satellite systems, downscaled CMAQ exposure concentration estimates, a spatiotemporal exposure concentration surface, a LUR model, and a combined LUR-kriging model. The mean and median $PM_{2.5}$ exposure concentrations were similar across methods (range of means: 11.4 to 12.2 $\mu g/m^3$), but the LUR models and one spatiotemporal model (geographically-weighted regression) produced higher variability than the other methods (IQRs range from 3.6 to 5.7 $\mu g/m^3$).

Epidemiologic study design influences the relevance and utility of exposure concentration estimation methods. Methods with high temporal resolution are preferable for short-term exposure studies even if spatial resolution is low, assuming the temporal variability at the site of data collection does not vary substantially across the study area. Fixed-site monitors, with temporal variability matching that of the health dataset, may be appropriate for this case, especially for $PM_{2.5}$ concentration, which tends to be less spatially variable than concentrations of $PM_{10-2.5}$ or UFP. Methods with high spatial resolution are preferable for long-term exposure studies where spatial contrasts are important. Methods that merge data from several sources, such as hybrid methods drawing from a combination of land use variables, satellite observations, CTM model output, and surface measurements, are designed to produce more spatial variability in the PM concentration surface. However, satellite data and CTM model output are not as readily available for $PM_{10-2.5}$ and UFP as they are for $PM_{2.5}$. Table 3-5 summarizes various exposure

concentration estimation methods used in PM epidemiologic studies, appropriate applications, and associated errors and uncertainties. In general, the methods listed in Table 3-5 that model spatial variability more accurately are often used in studies of health effects from long-term PM exposure, because uncertainties in spatial variability will have more of an influence on effect estimates from long-term exposure studies. Similarly, the methods that capture temporal variability are typically used in short-term PM exposure studies, because uncertainties in temporal variability will have more of an influence on effect estimates from short-term exposure studies.

**Table 3-5 Summary of exposure or exposure concentration estimation methods, their typical use in PM epidemiologic studies, and related errors and uncertainties.**

| Exposure Concentration Assignment Method | Description | Epidemiologic Application | Strengths | Limitations | Exposure Errors |
|---|---|---|---|---|---|
| *Measurement Methods* | | | | | |
| Fixed-site monitor [Section 3.3.1.1; Section 2.4.1; U.S. EPA (2009b)] | Typically, the nearest monitor to a receptor location; monitor type varies with particle size: $PM_{2.5}$: A FRM or FEM monitor located at a fixed location to measure ambient PM concentration; $PM_{10-2.5}$: A dichotomous FRM or FEM monitor located at a fixed location to measure ambient PM concentration, collocated $PM_{10}$ and $PM_{2.5}$ monitors used to calculate concentrations by differencing for a given location, or non-collocated $PM_{10}$ and $PM_{2.5}$ monitors used to calculate concentrations by differencing across a city or county; UFP: typically, a CPC to measure particle number concentration. | Short-term exposure studies: surrogate for ambient PM exposure concentration of a population within a city. Long-term exposure studies: surrogate for ambient PM exposure concentration to compare populations within a city or among multiple cities. | Ambient PM concentration measurements undergo rigorous quality assurance | Non-FRM and non-FEM optical instruments cannot be calibrated to ambient conditions, based on differences in size distributions and composition of calibration particles (e.g., Arizona road dust) and ambient PM; measurements of ambient PM concentration made at a fixed location may differ from an exposed individual's true exposure concentration, and no spatial variation is assumed; smaller particles (e.g., UFP) are more susceptible to evaporative losses. | Correlation between outdoor PM concentrations proximal to the receptors and ambient PM concentration measurements typically decreases with increasing distance from the monitor, especially for $PM_{10-2.5}$ and UFP, potentially leading simultaneously to decreased precision and to bias towards the null, as increased noise drives the slope towards zero; errors in $PM_{10-2.5}$ concentrations related to different flow rates used in $PM_{10}$ and $PM_{2.5}$ monitors for the differencing methods; errors in $PM_{10-2.5}$ concentrations due to differences in locations of $PM_{10}$ and $PM_{2.5}$ monitors when the instruments are not collocated. Potential for bias if ambient PM concentration at a receptor location is higher or lower than the ambient PM concentration measured at the monitor, especially for $PM_{10-2.5}$ and UFP; potential for imprecision from assumption of constant PM concentration within some radius of the monitor, especially for $PM_{10-2.5}$ and UFP; errors in $PM_{10-2.5}$ concentrations related to different flow rates used in $PM_{10}$ and $PM_{2.5}$ monitors for the differencing methods; errors in $PM_{10-2.5}$ concentrations due to differences in locations of $PM_{10}$ and $PM_{2.5}$ monitors when the instruments are not collocated. |

**Table 3-5 (Continued): Summary of exposure or exposure concentration estimation methods, their typical use in PM epidemiologic studies, and related errors and uncertainties.**

| Exposure Concentration Assignment Method | Description | Epidemiologic Application | Strengths | Limitations | Exposure Errors |
|---|---|---|---|---|---|
| Microenvironmental monitor (Section 3.3.1.2) | Typically located in an outdoor or indoor microenvironment to measure ambient PM concentration; $PM_{2.5}$: A FRM or FEM monitor located at a fixed location to measure ambient PM concentration; $PM_{10-2.5}$: A dichotomous FRM or FEM monitor located at a fixed location to measure ambient PM concentration, or collocated $PM_{10}$ and $PM_{2.5}$ monitors used to calculate concentrations by differencing for a given location; UFP: typically, a CPC to measure particle number concentration | Panel studies: PM exposure (e.g., personal or residential samples) within a geographic area | Ambient PM concentration measurements undergo rigorous quality assurance | Non-FRM and non-FEM optical instruments cannot be calibrated to ambient conditions, based on differences in size distributions and composition of calibration particles (e.g., Arizona road dust) and ambient PM; instrument expense may make it difficult to perform sampling simultaneously in multiple environments. | Nonambient PM exposure sampling may lead to bias if appropriate statistical methods are not used for handling biased data. |

**Table 3-5 (Continued): Summary of exposure or exposure concentration estimation methods, their typical use in PM epidemiologic studies, and related errors and uncertainties.**

| Exposure Concentration Assignment Method | Description | Epidemiologic Application | Strengths | Limitations | Exposure Errors |
|---|---|---|---|---|---|
| Active personal exposure monitor (Section 3.3.1.2) | Air is pulled through a pump and sampled for ambient PM concentration;<br>$PM_{2.5}$ or $PM_{10-2.5}$: air is typically directed through a collection filter on an impaction plate or past an optical detector; upstream hardware (e.g., cyclone) may be used for separating PM by specific size fractions;<br>UFP: typically, a CPC to measure particle number concentration; for BC, PM is typically measured with an aethalometer. | Panel studies: PM exposure (e.g., personal or residential samples) within a geographic area | PM and/or BC concentrations are obtained at the site of the exposed person | Non-FRM and non-FEM optical instruments cannot be calibrated to ambient conditions, based on differences in size distributions and composition of calibration particles (e.g., Arizona road dust) and ambient PM; some monitors can detect a minimum particle size of 0.1 µm and a few others can detect 0.25 µm, but the majority detect over the entire fine PM range; many monitors are noisy. | Nonambient PM exposure sampling may lead to bias if appropriate statistical methods are not used for handling biased data. |
| Passive personal exposure monitor (Section 3.3.1.2) | PM is captured on a treated substrate via passive exposure for a time period to measure a personal or area sample, and the substrate is analyzed by SEM; concentration is calculated based on a model of passive diffusion flux for $PM_{2.5}$, $PM_{10-2.5}$, or UFP. | Panel studies: ambient PM exposure within a city or among multiple cities | PM concentrations are obtained at the site of the exposed person | Long duration integrated sampling time (e.g., 7 days) does not allow for time-series analysis; diffusion-related losses to the passive sampler hardware have the potential to bias the concentration estimation based both on reduced particle counts and overestimation of flux to the sampling substrate. | Nonambient PM exposure sampling may lead to bias. |

**Table 3-5 (Continued): Summary of exposure or exposure concentration estimation methods, their typical use in PM epidemiologic studies, and related errors and uncertainties.**

| Exposure Concentration Assignment Method | Description | Epidemiologic Application | Strengths | Limitations | Exposure Errors |
|---|---|---|---|---|---|
| *Modeling Methods* | | | | | |
| Data averaging (Section 3.3.2.1) | Averaging across multiple monitors during the same time window and within a geographical area such as a city or county, typically using fixed-site monitoring data | Short-term exposure studies: surrogate for ambient PM exposure concentration of a population within a city | Ambient PM concentration measurements undergo rigorous quality assurance; averaging scheme designed for population or trend of interest | Non-FRM and non-FEM optical instruments cannot be calibrated to ambient conditions, based on differences in size distributions and composition of calibration particles (e.g., Arizona road dust) and ambient PM; measurements of ambient PM concentration made at a fixed location may differ from an exposed individual's true exposure concentration, and spatial variation is assumed to be well-represented by the averaging scheme. | Correlation between outdoor PM concentrations proximal to the receptors and ambient PM concentration measurements typically decreases with increasing distance from the monitor, especially for $PM_{10-2.5}$ and UFP, potentially leading simultaneously to decreased precision and to bias towards the null, as increased noise drives the slope towards zero. |
| | Spatial averaging (area averaging, population-weighted averaging), typically using fixed-site monitoring data | Long-term exposure studies: surrogate for ambient PM exposure concentration, usually within a city or geographic region | | | Potential for bias if ambient PM concentration at a receptor location is higher or lower than the spatial average, especially for $PM_{10-2.5}$ and UFP; potential for imprecision from assumption of constant PM concentration within some geographic area, especially for $PM_{10-2.5}$ and UFP. |

**Table 3-5 (Continued): Summary of exposure or exposure concentration estimation methods, their typical use in PM epidemiologic studies, and related errors and uncertainties.**

| Exposure Concentration Assignment Method | Description | Epidemiologic Application | Strengths | Limitations | Exposure Errors |
|---|---|---|---|---|---|
| Inverse distance weighting (Section 3.3.2.2) | Measured ambient PM concentrations are interpolated to estimate ambient PM concentration surfaces across regions; IDW uses an inverse function of distance to monitors | Long-term exposure studies: surrogate for ambient PM exposure concentration, usually within a city or geographic region | High spatial resolution | Over-smoothing based on assumption that ambient PM concentration is constant for a given distance from the source or based on smoothing function between monitors (which is more of an issue for $PM_{10-2.5}$ and UFP). | Potential for negative bias if ambient PM sources are not captured or PM concentration is overly smoothed; potential for positive bias if PM deposition or other loss processes; potential for imprecision from overly smoothed PM concentration. |
| Kriging (Section 3.3.2.2) | Measured ambient PM concentrations are interpolated to estimate ambient PM concentration surfaces across regions | Long-term exposure studies: surrogate for ambient PM exposure concentration, usually within a city or geographic region | High spatial resolution | Over-smoothing is possible based on smoothing function between monitors (which is more of an issue for $PM_{10-2.5}$ and UFP). | Potential for negative bias if ambient PM sources are not captured or PM concentration is overly smoothed; potential for positive bias if PM deposition or other loss processes; potential for imprecision from overly smoothed PM concentration. |
| Land use regression (Section 3.3.2.3) | Measured ambient PM concentrations are regressed on local variables (e.g., land use factors); the resulting model is used to estimate ambient PM concentrations at specific locations | Long-term exposure studies: surrogate for ambient PM exposure concentration, usually across a city but sometimes among multiple cities | High spatial resolution | Does not account for emission rates, dispersion, or atmospheric chemistry and may account for meteorology only in terms of wind speed and wind direction, depending on model formulation; has limited generalizability to other locations; uncertainties are highest where training monitors are sparse. | Potential for bias if grid is not finely resolved, if the model is misspecified, or if the model is applied to a location different from where the model was fit. |

**Table 3-5 (Continued): Summary of exposure or exposure concentration estimation methods, their typical use in PM epidemiologic studies, and related errors and uncertainties.**

| Exposure Concentration Assignment Method | Description | Epidemiologic Application | Strengths | Limitations | Exposure Errors |
|---|---|---|---|---|---|
| Spatiotemporal model (Section 3.3.2.3) | Measured ambient PM concentrations are modeled by a spatial average, spatially-varying covariates, and a spatiotemporal residual; the resulting model is used to estimate ambient PM concentrations at specific locations | Short-term and long-term exposure studies: surrogate for ambient PM exposure concentration, usually across a city but sometimes among multiple cities | High spatial resolution | Does not account for emission rates, dispersion, or atmospheric chemistry and may account for meteorology only in terms of wind speed and wind direction, depending on model formulation; has limited generalizability to other locations; uncertainties are highest where training monitors are sparse. | Potential for bias if grid is not finely resolved, if the model is misspecified, or if the model is applied to a location different from where the model was fit. |
| Chemical transport model (Section 3.3.2.4.1) | Grid-based ambient PM concentrations are estimated from emissions, meteorology, and atmospheric chemistry and physics | Short-term and long-term exposure studies: surrogate for ambient PM exposure concentration, sometimes within a city but more typically across a larger region | Strengths include accounting for emission rates, mixing height, atmospheric stability, meteorology, atmospheric chemistry, and complex terrain | Limited grid cell resolution (i.e., grid cell length scale is typically 4−36 km); spatial smoothing of local PM emissions sources; UFP not typically modeled; temporal emission allocations (e.g., by hour of weekday, by month, etc.) are generally the same over time. | Potential for bias when grid cells are too large to capture spatial variability of ambient PM exposures, especially for $PM_{10-2.5}$; bias in PM mass concentration and PM components related to underestimation of BC and OC. |
| Dispersion model (Section 3.3.2.4.2) | Ambient PM concentrations at specific locations are estimated from emissions, meteorology, and atmospheric physics | Short-term and long-term exposure studies: surrogate for ambient PM exposure concentration within a city or geographic region | High spatial and temporal resolution, accounts for atmospheric physics from local emission sources | Very limited representation of atmospheric chemistry or background PM concentrations; input emissions data are sometimes not available (e.g., roads where vehicle counts are not measured). | Potential for bias where the dispersion model does not capture boundary conditions and resulting fluid dynamics well (e.g., in large cities with urban topography affecting dispersion). |

**Table 3-5 (Continued): Summary of exposure or exposure concentration estimation methods, their typical use in PM epidemiologic studies, and related errors and uncertainties.**

| Exposure Concentration Assignment Method | Description | Epidemiologic Application | Strengths | Limitations | Exposure Errors |
|---|---|---|---|---|---|
| Hybrid approaches (Section 3.3.2.4.3) | Grid-based ambient PM concentrations are estimated from emissions, meteorology, and atmospheric chemistry and physics and bias corrected based on monitoring data | Short-term and long-term exposure studies: surrogate for ambient PM exposure concentration, sometimes within a city but more typically across a larger region | Strengths include accounting for emission rates, mixing height, atmospheric stability, meteorology, atmospheric chemistry, and complex terrain; bias correction improves model results, particularly where biases are large | Limited grid cell resolution (i.e., grid cell length scale is typically 4−36 km); resource-intensive; spatial smoothing of local PM emissions sources; UFP not typically modeled. | Although there is the potential for bias when grid cells are too large to capture spatial variability of ambient PM exposures (especially for $PM_{10-2.5}$; bias in PM mass concentration and PM components related to underestimation of BC and OC), fusing model results with monitoring data helps to minimize exposure errors. |
| Microenvironmental modeling [e.g., APEX, SHEDS (Section 3.3.4)] | Estimates distributions of micro-environmental PM concentrations, exposures, and doses for populations (e.g., census tracts) based on air quality data, demographic variables, and activity patterns | Short-term and long-term exposure studies; panel studies | Accounts for variability of PM exposures across large populations, accounts for different concentrations in different microenvironments, accounts for location-activity information | Models simulate individuals and their exposures; they do not model actual individuals but simulated representative individuals based on the population being modeled. | Potential for bias when the modeled distributions of ambient PM concentration, indoor:outdoor pollutant ratios, and time-activity patterns differ from the true distributions. |

**Table 3-5 (Continued): Summary of exposure or exposure concentration estimation methods, their typical use in PM epidemiologic studies, and related errors and uncertainties.**

| Exposure Concentration Assignment Method | Description | Epidemiologic Application | Strengths | Limitations | Exposure Errors |
|---|---|---|---|---|---|
| Satellite-based methods (Section 3.3.3) | Grid-based ambient PM concentrations are estimated from emissions, meteorology, and atmospheric chemistry and physics and bias corrected based on satellite data | Long-term exposure studies: surrogate for ambient PM exposure concentration, sometimes within a city but more typically across a larger region | Strengths include bias correction improves model results, particularly where biases are large | Limited temporal resolution (i.e., based on a daily observation); assume AOD is representative of ground-level $PM_{2.5}$ concentrations; algorithms converting AOD observations to $PM_{2.5}$ concentrations vary regionally; limited grid cell resolution (i.e., grid cell length scale is typically 1−36 km); spatial smoothing of local PM emissions sources; $PM_{10-2.5}$ and UFP not typically modeled. | Although there is the potential for bias when grid cells are too large to capture spatial variability of ambient PM exposures (especially for $PM_{10-2.5}$; bias in PM mass concentration and PM components related to underestimation of BC and OC), fusing model results with satellite data helps to minimize exposure errors. |

APEX = air pollutants exposure model; BC = black carbon; CPC = condensation particle counter; FEM = federal equivalent method; FRM = federal reference method; IDW = inverse distance weighting; SHEDS = stochastic human exposure and dose simulation; PM = particulate matter $PM_{2.5}$ = PM with a 50% cut point at 2.5 μm; $PM_{10-2.5}$ = PM fraction captured between 50% cut points of 10 μm and 2.5 μm; SEM = scanning electron microscopy; UFP = ultrafine PM.

## 3.4 Exposure Assessment and Interpretation of Epidemiologic Study Results

The exposure assignment methods discussed in Section 3.3 inform different PM-health relationships, depending on the method chosen. These relationships include those between ambient concentration and health effects, between exposure concentration and health effects, and between ambient exposure and health effects. The ambient exposure-health relationship is the main relationship of interest for the causal determinations in the ISA, and it can be evaluated using personal monitors, microenvironmental models, or ambient concentration as a surrogate for exposure (Table 3-5). Methods that estimate local exposure concentration, including spatial averaging, LUR, and emissions/transport models inform the exposure concentration-health relationship. Ambient concentration measured at an ambient monitor can be used directly to inform the ambient concentration-health relationship.

The following sections review the available literature to explore how the selection of an exposure metric may influence these relationships. The following discussion focuses on the relationships influencing exposure, such as those between ambient PM concentration and exposure to ambient PM (Section 3.4.1), factors contributing to error in estimating exposure to ambient PM (Section 3.4.2), and the influence of exposure errors on epidemiologic study results (Section 3.4.4). Additionally, this section explores copollutant relationships that may influence interpretation of the health effect estimates for ambient PM exposures (Section 3.4.3).

### 3.4.1 Relationships Influencing Exposure

This section builds upon discussions from the 2009 PM ISA (U.S. EPA, 2009b) about relationships between ambient PM measured outdoors, ambient PM infiltrating indoors, and resulting relationships between indoor and outdoor ambient PM concentrations and between personal exposure to ambient PM and ambient PM concentration. Summaries of relevant discussions from the 2009 PM ISA are included in Section 3.4.1.1, Section 3.4.1.2, and Section 3.4.1.3.

#### 3.4.1.1 Air Exchange Rate and Infiltration

When concentrations measured at an ambient monitor are used as a surrogate for $PM_{2.5}$, $PM_{10-2.5}$, or UFP exposure, the metric does not account for reduction in exposure concentration related to the process of infiltration indoors. The 2009 PM ISA (U.S. EPA, 2009b) describes how air exchange rate (AER) can influence the infiltration of PM into the building envelope. AER is the airflow into and out of a building and is represented by a in the conceptual model presented in Section 3.2.2. Several factors affect the AER, including weather conditions, building characteristics, and occupant behavior, resulting in

substantial spatial and temporal variations in AER. Deposition is dependent on PM size, where UFP loss can be expected to occur through Brownian diffusion, while $PM_{10-2.5}$ losses may occur through gravitational deposition or impaction. These phenomena were described in Sarnat et al. (2006a) and summarized in the 2009 PM ISA. New developments include characterizing infiltration of UFP, clarification on the factors influencing infiltration, and examination of air conditioning usage or AER as an effect modifier of $PM_{2.5}$ exposure for epidemiologic studies.

Field studies indicate that residential AER values vary by region and season, with substantial variability among different residences. Cao and Frey (2011) observed higher geometric mean AER in New York City (0.64 $hour^{-1}$), where housing stock tends to be older, compared with Harris County, TX (0.37 $hour^{-1}$) and a six-county region of central North Carolina (0.54 $hour^{-1}$). The RIOPA (Relationship Among Indoor, Outdoor, and Personal Air) study measured summer and winter AER in homes in three U.S. cities (Los Angeles, CA, Elizabeth, NJ, and Houston, TX). Median AER values were similar in Los Angeles and Elizabeth (0.87 $hour^{-1}$ and 0.88 $hour^{-1}$, respectively), but lower in Houston (0.47 $hour^{-1}$) (Yamamoto et al., 2010). Isaacs et al. (2013) analyzed seasonal RIOPA and DEARS data and found similar AER for the RIOPA cities and median AER of 0.92 $hour^{-1}$ in winter and 1.46 $hour^{-1}$ in summer. Summer AER was lower than winter AER in Elizabeth (0.88 $hour^{-1}$ vs. 1.07 $hour^{-1}$) and Houston (0.37 $hour^{-1}$ vs. 0.63 $hour^{-1}$). A similar seasonal difference was observed in Windsor, Ontario (0.14 $hour^{-1}$ vs. 0.3 $hour^{-1}$) (Wheeler et al., 2011). In contrast, Los Angeles AER values were higher in summer than winter (1.14 $hour^{-1}$ vs. 0.61 $hour^{-1}$). More prevalent use of open windows in Los Angeles and Detroit, where summertime tends to be less humid than in Elizabeth or Houston, may promote greater air exchange. These differences may grow smaller with the increased prevalence of air conditioning, because air conditioning usage is an important factor in infiltration (Allen et al., 2012). The higher winter AER values in the northern cities of Elizabeth and Windsor may be due to an increased "stack effect" resulting from indoor-outdoor temperature differential (Breen et al., 2014).

Between-city variability in residential building characteristics may explain heterogeneity in associations of $PM_{2.5}$ with risk estimates (Section 11.1.6.3.2). Baxter and Sacks (2014) explored this idea by performing *k*-means cluster analysis of factors related to AER, including percentage of homes with central air conditioning, mean year the home was built, and mean home size, from the American Housing Survey across 94 CBSAs across the U.S. Their analysis produced five clusters, labeled Clusters 1-5 by the study authors. Clusters 2 and 3 had high proportions of air conditioning (72% each), and those clusters primarily spanned the southern U.S. including the southeast and southwest. Homes in these clusters were built, on average, in 1989 and 1970. Cluster 1, which crossed the Northeast, Rust Belt, Pacific coast, and Denver, had slightly more than 1 quarter (27%) of homes with air conditioning, and had smaller homes on average (1,672 $ft^2$). Clusters 4 and 5 were primarily situated in the Northeast and Rust Belt, had air conditioning in 56 and 19% of homes, and were somewhat larger (2,098 $ft^2$ and 2,253 $ft^2$). In the latter three clusters, homes were built on average in 1954, 1959, and 1945. The results of Baxter and Sacks (2014) and Baxter et al. (2017), in a related study of short-term $PM_{2.5}$ exposure and mortality, support the

idea of a regional differences in building characteristics and health effects estimates based on north-south and east-west differences in housing clusters.

Vehicle AERs can be substantially higher than residential AERs, leading to rapid infiltration of on-road pollutants. Many factors affect vehicle AER, including whether windows are opened or closed, vehicle make and model, vehicle age, driving speed, and fan/recirculation setting on the vehicle ventilation system. The combined effect of these factors result in AERs that vary by more than two orders of magnitude, from less than 1 $hour^{-1}$ (approximately equivalent to a typical residential AER) to more than 100 $hour^{-1}$ (Hudda et al., 2011). In a model fit to AER measurements on 59 vehicles driven at three different speeds under recirculation conditions with closed windows, the most important variables were vehicle age, mileage, and speed, plus an adjustment for manufacturer (Fruin et al., 2011). Fan speed and vehicle shape were not influential variables.

More data have since been acquired to estimate $F_{inf}$ for UFP since the Sarnat et al. (2006a) study. Sarnat et al. (2006a) found that $F_{inf}$ reached a maximum for particles of 200 nm size and was sensitive to AER and PM composition. The smallest size they studied was 20 nm. Kearney et al. (2014) estimated daily $F_{inf}$ for $PM_1$, $PM_{2.5-1}$, and UFP (NC estimated by the authors to have 80% smaller than 100 nm) in Edmonton, Ontario. They studied conditions in winter and summer and observed winter-time median $F_{inf}$ of 0.45 for $PM_1$ (based on the $SO_4^{2-}$ method) and of 0.19 for UFP (based on P-TRAK portable sampler measurements), a 58% reduction. During the summer, median $F_{inf}$ was 0.79 for $PM_1$ and 0.51 for UFP, a 35% reduction. In addition to the influence of season, Kearney et al. (2014) also tested building age and ventilation characteristics and found that building age, airflow characteristics in the home, temperature differential, and wind speed influenced $F_{inf}$ for $PM_1$ in winter, while furnace operation and wind speed influenced $F_{inf}$ for UFP in winter. For summer, only wind speed influenced $F_{inf}$ for $PM_1$, while portable air cleaner operation and window opening influenced $F_{inf}$ for UFP. Rim et al. (2010) focused on UFP smaller than 100 nm and were able to measure particles as small as 4.4 nm (under open window conditions) and 9 nm (under closed window conditions) in their study of $F_{inf}$ using an SMPS. For open window conditions, $F_{inf} = 0.08$ for particles in the 4.4–5.1 nm bin. For closed window conditions, $F_{inf} = 0.03$ for the 9–11 nm bin. For the 55–64 nm bin, $F_{inf}$ was 0.16 for closed windows and 0.47 for open windows. The Rim et al. (2010) study also compared the $C_{in}/C_{out}$ ratio with $F_{inf}$. Unlike for $PM_{2.5}$ and $PM_{10-2.5}$, the $C_{in}/C_{out}$ ratio was very close in value to $F_{inf}$ for UFP. These findings imply that very little PM in the smallest size fractions infiltrates the building envelope, suggesting that large errors would occur from assuming that concentrations measured at an ambient monitor were representative of indoor exposure to ambient UFP, especially as the particle size decreased.

Indoor air filtration using high-efficiency particulate air (HEPA) filters can reduce $F_{inf}$ as well as indoor total and ambient $PM_{2.5}$ concentrations. Allen et al. (2011) conducted an intervention study by temporarily installing HEPA filters in 25 homes in British Columbia, Canada during winter and early spring. Indoor $PM_{2.5}$ concentrations were 59% lower on average during HEPA filter operation (4.6 vs. 11.2 $\mu g/m^3$). Reductions of similar magnitude were observed for outdoor-generated $PM_{2.5}$

(1.5 vs. 3.5 μg/m$^3$). Allen et al. (2011) estimated $F_{inf}$ using the recursive method of Allen et al. (2003) and found that the average infiltration of $PM_{2.5}$ was reduced by 41% (0.20 vs. 0.34). These studies show a consistent effect of HEPA filtration in reducing $PM_{2.5}$ infiltration.

Several recent studies suggest that air conditioning may modify the association between $PM_{2.5}$ and health effects. Allen et al. (2012) used $PM_{2.5}$ and questionnaire data from the MESA-Air study to model $F_{inf}$ as a function of air conditioning and heating use, window opening, and window insulation. During the summer, central air conditioning usage was the most important factor in the model, accounting for 80% of the overall model variability (model $R^2 = 0.70$). During the winter, the most important factor was 2-week average outdoor temperature, which accounted for 45% of the overall model variability (model $R^2 = 0.49$). These results suggest that the variability in $PM_{2.5}$ infiltration within and between cities may account for increased variability in estimation of $PM_{2.5}$ exposure and hence attenuation of the health effect estimate. Hodas et al. (2012) considered sensitivity of $F_{inf}$ to $PM_{2.5}$ mass concentration, $PM_{2.5}$ component concentration, proximity to roadways, and income. Generally speaking, $F_{inf}$ was higher when calculated for $PM_{2.5}$ mass concentration rather than individual components. $F_{inf}$ was higher for both those living near roadways and for AER of 0.90 hour$^{-1}$, which was identified as the "typical" AER for low income homes compared with the general population. Hodas et al. (2012) suggested that variation in F may account for exposure misclassification in cases where variability in AER leads to assignment of incorrect F and for effect modification when conditions such as source proximity and poverty influence F.

Based on results of studies showing how $F_{inf}$ varies under different conditions, Allen et al. (2012) suggested that infiltration could modify the health effect of $PM_{2.5}$ exposure; this idea was explored in other studies. Bell et al. (2009) tested if air conditioning prevalence (i.e., the proportion of homes with air conditioning in a given community as indicated by the American Housing Survey) modified the effect of $PM_{2.5}$ exposure concentration on cardiovascular and respiratory hospital admissions (HA) and of $PM_{10}$ on mortality. Over the course of a year they observed decreases of 30% for the effect of short-term $PM_{10}$ exposure on mortality and of 34% for the effect of short-term $PM_{2.5}$ exposure on cardiovascular HA when any air conditioning was in use. They observed an overall 45% increase in the effect of $PM_{2.5}$ on respiratory HA for those who use air conditioning, but a break-down of their data showed that there was a 75% decrease in effect of $PM_{2.5}$ on respiratory HA during the summer when air conditioning use would be most prevalent. Sarnat et al. (2013a) also explored how AER can be a modifier of the effect of $PM_{2.5}$, $NO_X$, and CO related to asthma ED visits in Atlanta neighborhoods. Parsing their data by low and high AER (0.25/hour threshold) and poverty level (8.5% threshold), Sarnat et al. (2013a) observed that the majority of locations with high levels of poverty also had high AER. They attributed this observation to old, drafty housing being more prevalent among those in poverty. Larger effect estimates were observed among those with high poverty and low AER, however. When effect modification was tested using an interaction term, a negative effect on ED asthma visits was observed despite increased $PM_{2.5}$ and AER being associated with increased ED visits. These results indicate that air conditioning may modify associations between $PM_{2.5}$ and health effects, but the results are not entirely consistent.

Many of the newer studies of PM infiltration focused on characterizing infiltration of UFP, clarification on the factors influencing infiltration, and examination of air conditioning usage or AER as an effect modifier of $PM_{2.5}$ exposure. UFP infiltration was found to decrease with decreasing particle size, likely due to particle diffusion to surfaces. Many new studies noted differences in infiltration for seasons or between northern and southern cities. Areas with prevalent air conditioning usage tended to have lower infiltration compared with areas where window opening is prevalent. Indoor-outdoor temperature gradients also likely influenced PM infiltration, with particles naturally following the warm-cold gradient. Some recent studies found that air conditioning may also modify the effect of short-term $PM_{2.5}$ exposure and health effects.

### 3.4.1.2 Indoor–Outdoor Concentration Relationships

The 2009 PM ISA (U.S. EPA, 2009b) largely focused on infiltration of PM in the $PM_{2.5}$ and $PM_{10-2.5}$ size ranges, finding that infiltration of PM indoors decreased with increasing particle size. This section builds on the literature review from the 2009 PM ISA with a focus on relationships between indoor and local outdoor PM concentrations in different size fractions, particularly $PM_{2.5}$ and UFP. Most of the studies published since the 2009 PM ISA that evaluated indoor-outdoor PM relationships were conducted outside the U.S., including studies in Europe, Canada, Mexico, South America, the Middle East, and Asia. Since PM levels, sources, and composition are likely to differ substantially in some areas from those typically encountered in the U.S., this section focuses on North American and European indoor-outdoor studies.

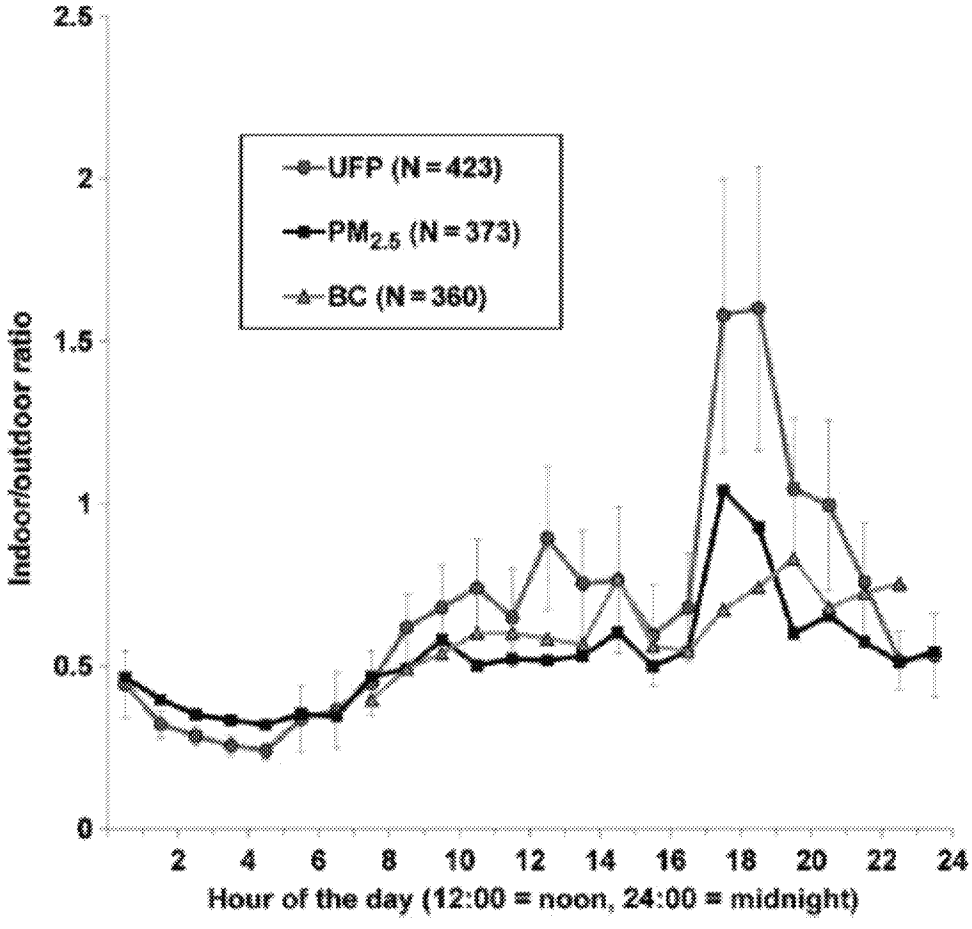
Recent literature has added data to the characterization of indoor-outdoor relationships across the $PM_{2.5}$ and $PM_{10-2.5}$ size fractions. A multicity study in Europe compared indoor and outdoor residential 24-hour average concentrations for NC (7−3,000 nm), $PM_{2.5}$, and $PM_{10-2.5}$ at 152 homes in Helsinki (Finland), Athens (Greece), Amsterdam (the Netherlands), and Birmingham (U.K.) (Hoek et al., 2008b). Median indoor-outdoor correlations for $PM_{10-2.5}$ were the lowest of the three PM metrics in all cities, ranging from 0.10−0.39. In Helsinki and Amsterdam, NC indoor-outdoor correlations were lower than $PM_{2.5}$ correlations (0.41 vs. 0.74 and 0.58 vs. 0.85, respectively), while in Athens and Birmingham, NC correlations were higher (0.80 vs. 0.63; 0.50 vs. 0.35). A common indoor source, gas cooking, was prevalent in both Amsterdam and Birmingham, cities with differing correlation magnitude, and so is unlikely to explain city-to-city differences in correlations. Consistent with observed low correlations, the regression slope of indoor on outdoor concentrations (a measure of infiltration, with a slope less than one indicating less infiltration) was lower for $PM_{10-2.5}$ than the other two PM metrics, ranging from 0.11−0.16. NC slopes ranged from 0.19−0.42 and were lower than $PM_{2.5}$ slopes (range: 0.39−0.48) in Amsterdam, Birmingham, and Helsinki, while the two slopes were roughly equivalent in Athens. Again, infiltration slope results were generally consistent with correlation results, being either both high or both low in a particular city. Buonanno et al. (2013a) reported I/O and the ratio of indoor to fixed-site monitors for three schools in Cassini, Italy and found I/O ranged from 0.63−0.74 while the indoor to fixed-site ratio

ranged from 0.47−1.53. These values are much higher than those reported in the Hoek et al. (2008b) study. Another important finding is that $PM_{10-2.5}$ exhibited the lowest infiltration and indoor-outdoor correlation of the three metrics, with NC and $PM_{2.5}$ infiltration behavior similar to one another. Semmens et al. (2015) measured NC in various size fractions ranging from 0.3−10 μm and found that correlations between indoor $PM_{2.5}$ and various NC size fractions were very high for NC less than 1 μm in size (0.94 and 0.93 for NC 0.3−0.49 μm and 0.5−0.99 μm, respectively). Correlations with $PM_{2.5}$ decreased monotonically for larger NC size fractions, with $PM_{2.5}$–$PM_{10-2.5}$ correlations of 0.46 for NC 2.5−4.99 μm and 0.35 for NC 5.0−9.99 μm. Correlations among indoor NC size fractions were highest for adjacent bins. Collectively, these results indicate that differences in source patterns, spatial concentration heterogeneity, housing stock, meteorology, and other factors contribute to different indoor-outdoor relationships in different urban areas, particularly for NC and $PM_{2.5}$.

Results for indoor-outdoor relationships for $PM_{2.5}$ concentration were not consistent across studies of the effect of season. Several single-city studies in the U.S. and Canada have evaluated indoor-outdoor relationships by season. For example, in Boston, median residential indoor-outdoor slopes for 24-hour average $PM_{2.5}$ were higher in summer than winter (0.74 vs. 0.53) for a panel of 25 participants studied in 2000 (Brown et al., 2008). Hsu et al. (2012) reported correlations between indoor and outdoor (outside residence and fixed-site monitors) concentrations of $PM_{10-2.5}$ and $PM_{2.5}$ in New York City, NY and Seattle, WA. For $PM_{10-2.5}$ in New York City (correlations not reported for Seattle), Spearman $R = 0.20$ for indoor-outdoor and 0.08 for indoor-fixed-site during the summer and Spearman $R = -0.12$ and −0.07 for indoor-outdoor and indoor-fixed-site during the winter. For $PM_{2.5}$ in New York City, Spearman $R = 0.44$ for both indoor-outdoor and indoor-fixed-site in winter and Spearman $R = 0.57$ and 0.53 for indoor-outdoor and indoor-fixed-site in summer. Hochstetler et al. (2011) measured $PM_{2.5}$, EC, and NC inside and outside three public schools in Cincinnati, OH and observed a lower slope and $R^2$ for $PM_{2.5}$ (I/O slope = 0.24, $R^2 = 0.08$), compared with EC (I/O slope = 0.44, $R^2 = 0.66$) and NC (I/O slope = 0.68, $R^2 = 0.72$). In Windsor, Ontario, Kearney et al. (2011) calculated the indoor-outdoor ratio (I/O) for UFP (20−100 nm), and found wide variation with median I/O of 0.19 (95th percentile: 0.64) and 0.27 (95th percentile: 0.61) for summer measurements for 2005 and 2006, respectively, and 0.25 (95th percentile: 0.45) for winter, 2006 measurements. Kearney et al. (2011) based these numbers on nighttime measurements, when it was assumed that there were no indoor sources of UFP so that I/O approximates $F_{inf}$; I/O estimates based on recursive and censoring models produced similar results. Daily I/O (not slopes) in Windsor were similar for $PM_{2.5}$ (0.5), BC (0.45), and 20−1,000 nm NC (0.55) at approximately 90 residences, averaging across summer and winter sampling seasons (Wheeler et al., 2011). Hourly I/O for NC were much higher during dinnertime (approximately 1.5), indicating indoor NC sources from cooking (Figure 3-2); this also contributed to a higher daily ratio relative to the other PM metrics. For $PM_{10-2.5}$ in Regina, Saskatchewan, 5-day geometric mean concentrations were lower indoors than outdoors during summer (4.3 vs. 8.8 $\mu g/m^3$) in a set of 100 residences, but the opposite was true for a set of 79 residences during winter, with higher indoor concentrations (3.7 vs. 2.5 $\mu g/m^3$). The spatial coefficient of variation for outdoor $PM_{10-2.5}$ concentrations was higher in winter than in summer.

Variation in indoor-outdoor relationships among different studies for warm and cold months may relate to different contributions from indoor sources, such as cooking and heating, between cities.

Time of day also influences I/O ratios, as shown in Figure 3-3 for data reported by Wheeler et al. (2011). In addition, Semmens et al. (2015) studied residences relying mainly on wood stoves for heating and found that I/O ratios were approximately 1.0−1.2 (indicating indoor sources) during daytime hours (6 a.m.–10 p.m.), indicating the wood stove or other indoor sources were contributing to indoor PM. Overnight (10 p.m.–6 a.m.) ratios were approximately 0.6. The relatively lower overnight I/O supports the finding that indoor sources were driving the high I/O values during the day.

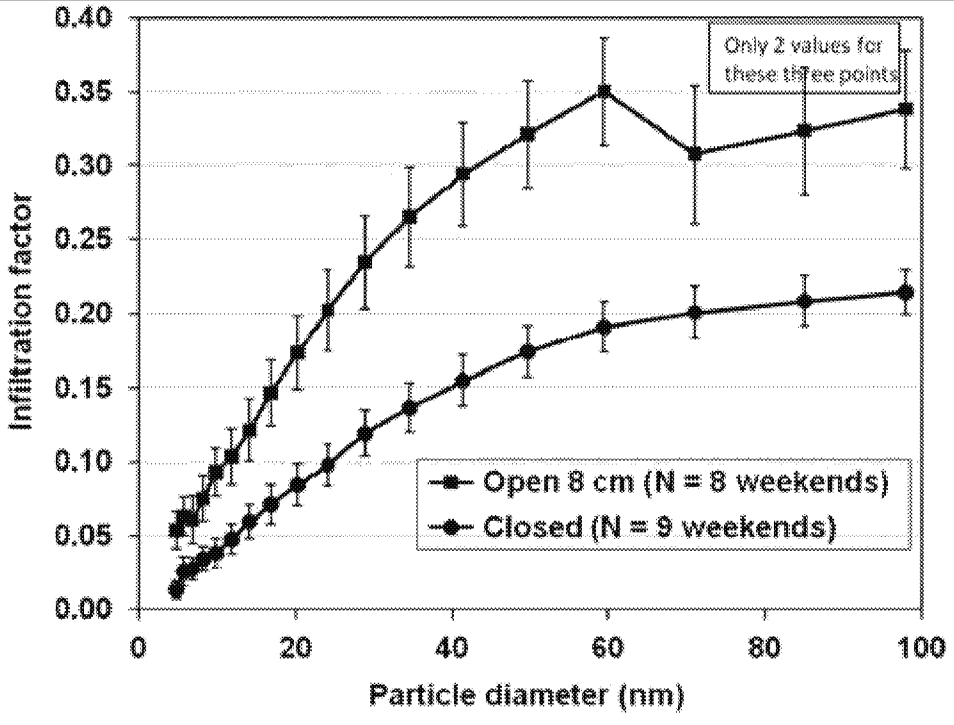



Note: Standard errors are only shown for the I/O for UFP. This figure was reproduced from Wheeler et al. (2011). The figure shows how the indoor-outdoor ratios change with hour of day for UFP, $PM_{2.5}$, and BC. Each type of PM has a peak indoor-outdoor ratio between 17:00 and 20:00. However, the peak indoor-outdoor ratio is much higher for UFP than for $PM_{2.5}$, which is slightly higher than for BC.

Source: Permission pending Wheeler et al. (2011).

**Figure 3-2 Indoor-outdoor ratios for UFP, $PM_{2.5}$, and BC measured at 90 residences.**

New research on UFP I/O suggest that I/O decreases with decreasing particle size within the ultrafine size range. Indoor-outdoor ratios were calculated for a manufactured house located on the National Institute for Standards and Technology (NIST) campus in Gaithersburg, MD to characterize infiltration to test how I/O varies across UFP size (Wallace and Ott, 2011). I/O generally increased with increasing UFP size (up through 100 nm) for both open and closed window conditions (Figure 3-3). Open window I/O was always higher and had greater variability than closed window I/O. This pattern is consistent with observations by Sarnat et al. (2006a) presented in the 2009 PM ISA (U.S. EPA, 2009b) in which $F_{inf}$ increases with increasing particle size up to about 100 nm. Above 200 nm, Sarnat et al. (2006a) reported that $F_{inf}$ declined with increasing particle size up to 8 μm. Across all experiments, Wallace and Ott (2011) estimated that ambient UFP exposure was responsible for 36% of total UFP exposure and that the contribution of outdoor UFP exposure to total UFP exposure would likely increase in urban environments.




Source: Permission pending Wallace and Ott (2011).

**Figure 3-3 Indoor-outdoor ratios for UFP size obtained in a test house on the National Institute for Standards and Technology (NIST) facility for open and closed window conditions.**

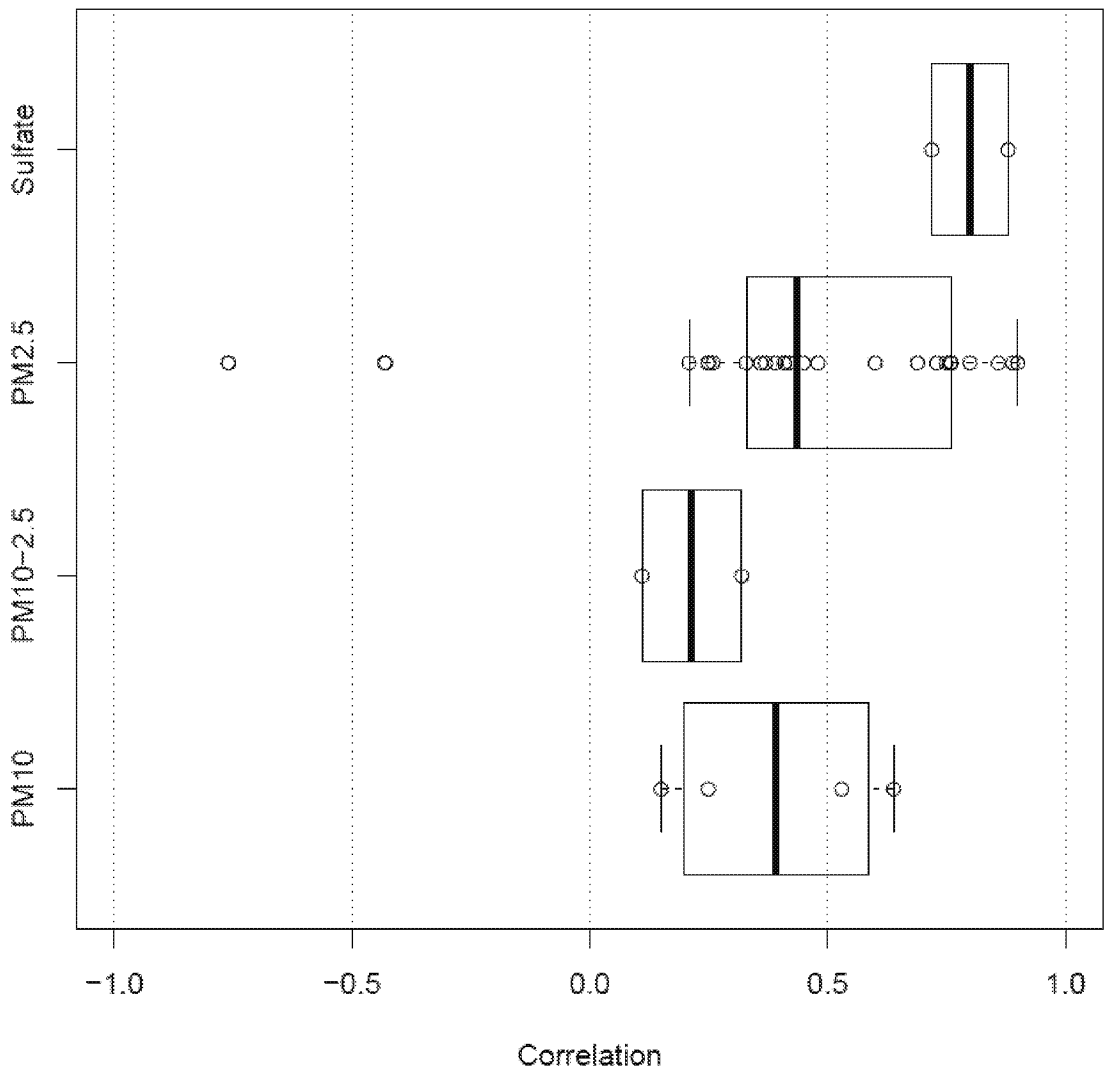
Recent studies reinforce previous conclusions that I/indoor-outdoor relationships are sensitive to particle size, with I/O typically decreasing in the $PM_{10-2.5}$ range. New studies add to the literature base for UFP, where I/O was found to decrease with decreasing particle size. UFP movement is more influenced by Brownian diffusion than are larger particles, which likely caused more UFP to diffuse to building surfaces instead of being transported indoors. Additional studies added to the characterization of indoor-outdoor relationships for different seasons and times of day. For most studies, I/O was higher during summer than winter and during daytime compared with nighttime.

#### 3.4.1.3 Personal–Ambient Concentration Relationships

The new literature on personal-ambient relationships adds to findings from the 2009 PM ISA (U.S. EPA, 2009b), in which moderate correlations (0.3−0.7) were observed with median personal-ambient slope slightly higher than 0.5. The general understanding of these relationships is unchanged since the 2009 PM ISA. As with the previous section on indoor-outdoor relationships (Section 3.4.2), many of the studies published since the 2009 PM ISA that evaluated personal-ambient PM relationships were conducted outside the U.S., including studies in Europe, Mexico, South America, the Middle East, and Asia. Since PM levels, sources, and composition are likely to differ substantially in some areas from those typically encountered in the U.S., this section focuses on North American and European personal-ambient studies.

High correlations suggest that ambient concentrations are a good surrogate for personal exposure, while low correlations indicate exposure measurement error when using ambient concentration to represent personal exposure. Several studies, many of which were available at the time of the 2009 PM ISA (U.S. EPA, 2009b), have evaluated relationships between personal exposure and ambient PM concentrations in various U.S. cities, including: Baltimore, MD; Boston, MA; Chapel Hill, NC; Detroit, MI; and Steubenville, OH (Meng et al., 2012; Brown et al., 2009; Williams et al., 2008; Sarnat et al., 2006b; Koutrakis et al., 2005; Sarnat et al., 2005; Chang et al., 2000; Sarnat et al., 2000). These studies all evaluated 24-hour average exposures, except for Chang et al. (2000), which evaluated hourly exposures in a variety of microenvironments (e.g., indoor-home, indoor-other, outdoor-near-road, in-vehicle). Figure 3-4 shows personal-ambient correlations reported for Baltimore in Chang et al. (2000) and Sarnat et al. (2000) and New York City (Hsu et al., 2012). Both Baltimore studies evaluated $PM_{2.5}$, and Sarnat et al. (2000) reported personal-ambient correlations for $PM_{10}$, $PM_{10-2.5}$, and $SO_4^{2-}$. Hsu et al. (2012) also reported personal-ambient correlations for $PM_{10}$. Correlations ranged widely for $PM_{2.5}$, with a median of approximately 0.4 and an IQR of 0.3−0.7. $PM_{10}$ correlations were similar to those for $PM_{2.5}$, while $PM_{10-2.5}$ correlations were somewhat lower, suggesting factors such as spatial variability and differential infiltration affect exposure to ambient $PM_{10-2.5}$. These results also suggest that $PM_{10}$ was comprised primarily of $PM_{2.5}$ in these samples. Sulfate correlations were higher than those for $PM_{2.5}$. The recent findings of Hsu et al. (2012), in conjunction with older studies in the literature, indicate that a

greater portion of the variability in personal exposures is explained by variability in ambient PM for $PM_{2.5}$ and sulfate in $PM_{2.5}$, which tend to have lower spatial variability than $PM_{10-2.5}$ and UFP.

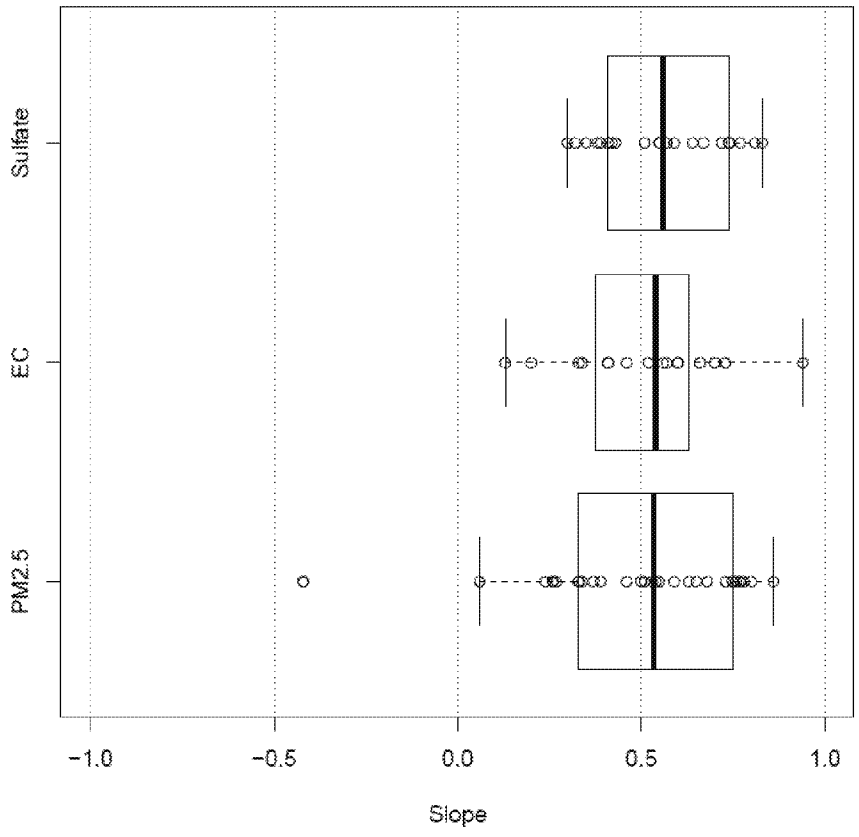



Source: Permission pending, Hsu et al. (2012); Chang et al. (2000); Sarnat et al. (2000).

**Figure 3-4 Correlations between personal exposure and ambient PM concentration in Baltimore, MD.**

Regressing personal exposure on ambient PM concentration yields a slope factor expressing the fraction of personal exposure from ambient PM. Figure 3-5 presents personal-ambient slopes (i.e., the ratio of total personal exposure to ambient concentration) from studies in the four cities listed previously (Meng et al., 2012; Brown et al., 2009; Sarnat et al., 2006b; Koutrakis et al., 2005; Sarnat et al., 2005). Several of these studies evaluated EC and $SO_4^{2-}$ in addition to $PM_{2.5}$. Median slopes for $PM_{2.5}$, EC, and $SO_4^{2-}$ were between 0.5 and 0.6. The wide variability in personal-ambient slopes is likely due in part to the study design, which evaluated personal exposure in different seasons and with different building ventilation conditions (e.g., closed vs. open windows). The variability may have also been attributed to variation in penetration and deposition for the components and houses. Ryan et al. (2015a) and Brokamp et al. (2015) analyzed concentration data from outdoor concentrations (outside residence) and total personal exposure samples for $PM_{2.5}$ mass and 24 $PM_{2.5}$ trace metals (Ag, Al, As, Ba, Br, Ca, Cl, Cr, Cu, Fe, K, Mn, Ni, Pb, S, Sb, Se, Si, Sn, Sr, Ti, V, Zn, Zr) from the RIOPA study of homes in Los Angeles, CA, Houston, TX, and Elizabeth, NJ. They presented correlation and outdoor-personal ratios (O/P) for each $PM_{2.5}$ component. Correlations of Spearman $R > 0.8$ were reported for S and V, while Spearman $R < 0.4$ was reported for Ag, Al, As, Ba, Ca, Cl, Cr, Cu, Fe, K, Mn, Ni, Sb, Si, Sr, Ti, Zn, Zr, and for $PM_{2.5}$ mass. Median O/P > 1 was observed for As, Br, Sb, Se, and V and O/P < 1 for $PM_{2.5}$ and the other components. The results for $PM_{2.5}$ and $PM_{2.5}$–S contrast those presented in Figure 3-5. Data were unavailable for $PM_{10-2.5}$ or UFP in these studies. These findings indicate that variability in the personal-ambient slope reflects differences in ventilation and other localized conditions for $PM_{2.5}$ mass concentration, which is not very sensitive to $PM_{2.5}$ composition.

New studies agree with the previously published literature on personal-ambient relationships. Studies have examined personal-ambient correlations for different PM size fractions and found that a greater portion of the variability in personal exposures is explained by variability in ambient PM for $PM_{2.5}$ and sulfate in $PM_{2.5}$, compared with $PM_{10-2.5}$, which tends to have greater spatial variability than $PM_{2.5}$. Median personal-ambient slopes are generally slightly greater than 0.5, and they likely reflect differences in residential ventilation, time-activity patterns (Section 3.4.2.1), and other localized conditions.




Source: Permission pending, Meng et al. (2012); Brown et al. (2009); Sarnat et al. (2006b); Koutrakis et al. (2005); Sarnat et al. (2005).

**Figure 3-5 Slopes of the relationship between personal exposure and ambient PM concentration in four U.S. cities.**

### 3.4.2 Factors Contributing to Error in Estimating Exposure to PM

This section builds upon discussions from the 2009 PM ISA (U.S. EPA, 2009b) about factors having the potential to cause error in exposure concentration estimates. Time-activity patterns, spatial variability, instrument error, and model accuracy and precision are discussed below, because these topics were frequently examined in exposure measurement error discussions. Summaries of each factor's discussion from the 2009 PM ISA are included in Section 3.4.2.1, Section 3.4.2.2, Section 3.4.2.3, and Section 3.4.2.4.

### 3.4.2.1 Time–Activity Patterns

The 2009 PM ISA (U.S. EPA, 2009b) reviewed time-activity behaviors among the population and how time spent in different locations varies among age groups. Recent additions have been made to time-activity databases, and technological advances in geographic positioning system (GPS) technologies have also expanded the information base regarding time-activity. Such new tools have enabled examination of factors that influence time-activity patterns and errors in those relationships.

Updated data are available from the Consolidated Human Activity Database (CHAD) to compare time-activity among different population strata for 25,431 individuals (Isaacs, 2014). Across the population, 75% of time is spent indoors at the place of residence; 5.5% is spent in transit; 16% indoors at work, school, or other locations; and 2.9% outdoors (Table 3-6). Substantially more time (82 and 83%) is spent indoors at home for children younger than 6 years and for adults older than 64 years, while teens ages 12−19 years and adults 20−64 years spent the least amount of time indoors at home (72 and 71%, respectively). Similarly, young children spent the least amount of time in transit (4.0%), while adults 20−64 years spent the most time in transit (6.9%). Adults 20−64 also spent the largest proportion of the day outdoors (3.4%), while older adults spent the least amount of time outdoors (2.2%). Young children ages 0−5 years and children ages 6−11 years spent less time outdoors than adults (2.4 and 3.0%, respectively). When comparing time-activity data across race (Table 3-7), Hispanic study participants spent slightly more time indoors at home than average (78%), while White study participants spent the most time outdoors (3.3%) compared with Asian (2.0%), Black (2.1%), and Hispanic (2.3%) participants. Males spent more time outdoors compared with females (3.6 vs. 2.2%) (Table 3-8), and adults 20−64 years with low and high education both spent less times indoors at home (74 and 70%, respectively), more time indoors at work/school/other (16 and 19%), and more time outdoors (3.7 and 3.5%) compared with the 20−64 year-old adult population (3.4%) (Table 3-9). It is possible that missing education data corresponded with lower time spent outdoors. It was most surprising to find that children spent less time outdoors than adults, while sex-specific differences in time-activity data were anticipated.

Recent studies have focused on the use of GPS technologies, such as in smartphones, to develop detailed time-activity pattern data. For example, Glasgow et al. (2014) analyzed the frequency of Android-based smartphones in recording positional data among a panel of study participants and found that on average 74% of the data were collected over intervals shorter than 5 min, which is a marked improvement over many time-activity studies using diaries.

**Table 3-6 Total and age-stratified time activity data from the Consolidated Human Activity Database.**

| Location Type | All | 0–5 yr | 6–11 yr | 12–19 yr | 0–19 yr | 20–64 yr | 65+ yr |
|---|---|---|---|---|---|---|---|
| Indoor-residential | 75.1% | 82.0% | 74.4% | 71.6% | 76.2% | 71.4% | 82.9% |
| Transit | 5.53% | 3.96% | 4.29% | 5.13% | 4.42% | 6.92% | 5.14% |
| Indoor-work/school/other | 15.5% | 10.1% | 16.7% | 19.9% | 15.3% | 17.9% | 8.71% |
| Outdoor | 2.87% | 2.35% | 2.96% | 2.53% | 2.62% | 3.39% | 2.18% |
| Uncertain or missing | 0.97% | 1.59% | 1.65% | 0.85% | 1.40% | 0.48% | 1.05% |

**Table 3-7 Total and race/ethnicity-stratified time activity data from the Consolidated Human Activity Database.**

| Location Type | All | Asian | Black | Hispanic | White |
|---|---|---|---|---|---|
| Indoor-residential | 75.1% | 75.3% | 74.8% | 78.4% | 74.8% |
| Transit | 5.53% | 5.01% | 5.25% | 5.05% | 5.54% |
| Indoor-work/school/other | 15.5% | 16.3% | 16.6% | 13.4% | 15.0% |
| Outdoor | 2.87% | 2.02% | 2.09% | 2.34% | 3.30% |
| Uncertain or missing | 0.97% | 1.42% | 1.26% | 0.84% | 1.45% |

**Table 3-8 Total and sex-stratified time activity data from the Consolidated Human Activity Database.**

| Location Type | All | Female | Male |
|---|---|---|---|
| Indoor-residential | 75.1% | 76.6% | 73.4% |
| Transit | 5.53% | 5.47% | 5.60% |
| Indoor-work/school/other | 15.5% | 14.8% | 16.4% |
| Outdoor | 2.87% | 2.21% | 3.64% |
| Uncertain or missing | 0.97% | 0.92% | 1.04% |

**Table 3-9 Total and education-stratified time activity data from the Consolidated Human Activity Database, among adults 20–64 years.**

| Location Type | All 20–64 yr | Low Education | High Education |
|---|---|---|---|
| Indoor-residential | 71.4% | 73.7% | 70.0% |
| Transit | 6.92% | 6.42% | 7.12% |
| Indoor-work/school/other | 17.9% | 16.0% | 19.1% |
| Outdoor | 3.39% | 3.73% | 3.52% |
| Uncertain or missing | 0.48% | 0.22% | 0.27% |

Positional errors are a concern for GIS and GPS-based technologies. Several studies found that median positional errors based on smartphones were less than 26 m (Ganguly et al., 2015; Lane et al., 2013; Wu et al., 2010). Glasgow et al. (2014) observed much larger errors, with an overall median positional accuracy of 342 m and a range from 98 to 1,169 m using an Android-based smartphone, while Wu et al. (2010) observed much smaller errors when comparing two smartphones with three other GPS technologies. To test the impact of the positional errors on concentration estimates used in exposure assessment studies, Ganguly et al. (2015) compared R-LINE modeled residential $PM_{2.5}$ concentrations when the positions were estimated with GIS or GPS over buffers of 0–100 m, 100–200 m, 200–500 m, and >500 m. Median concentration measurement errors were 5% or less for each buffer for annual average concentrations and 6% or less for 24-hour max concentrations. Average errors were 10% or less for each buffer for both annual average and 24-hour max concentrations.

Survey tools to assess time-activity may be subject to recall error among the subjects. Spalt et al. (2015) administered a survey to all participants in the Multi-Ethnic Study of Atherosclerosis (MESA) Air Study to ascertain information about time spent indoors and outdoors at home, at work/volunteer/school, in transit, or in other locations. A subset of the study population was asked to complete a time-activity diary as well. Correlation for indoor locations was Spearman $R = 0.63$ for home, Spearman $R = 0.73$ for work/volunteer/school, and Spearman $R = 0.20$ for other locations. Correlation for outdoor locations was much lower, with Spearman $R = 0.14$ at home, Spearman $R = 0.20$ for work/volunteer/school, and Spearman $R = 0.10$ for other locations. In transit, Spearman $R = 0.39$. These results suggest that study participants have better recall of the times spent inside their home or work/volunteer/school compared to other activities, because time spent at home or at work/volunteer/school tends to occur at routine times.

Excluding time-activity patterns from exposure studies may lead to bias and uncertainty in the exposure estimate. Nyhan et al. (2018) combined GPS records from 407,435 individuals in the metropolitan Boston, MA area with a hybrid model using land use regression and satellite data to predict $PM_{2.5}$ concentration on an hourly basis. They compared the time-activity-based model with one that used the daily average $PM_{2.5}$ concentration (also based on the hybrid LUR-satellite model) at location of resident for each participant and found that the residence-based exposure model produced predictions that were 9% lower than the model accounting for time-activity when averaging the results over a year. This suggests that omission of time-activity data may lead to underestimation of the exposure.

Residential mobility is one factor leading to error in estimating exposure for long-term exposure studies. Using a single address to represent exposure concentration over a period of several years may result in either under- or over-estimating exposure during the study period. For example, Brokamp et al. (2015) analyzed residential mobility for a cohort of children over the first seven years of life in Cincinnati, OH and found that 54% of the children changed residential address during that time, resulting in a 4.4% decrease in the cohort's average traffic-related air pollution exposure concentration (defined as BC estimates from an LUR model). They also noted that if the birth address is used for exposure estimation during the entire study period, exposure misclassification is increased for those that move earlier (due to more years at the incorrect address) or are more highly exposed (due to a greater likelihood of moving). An epidemiologic study of asthma incidence at age seven showed that not accounting for residential mobility resulted in bias toward the null.

Recognizing that the CHAD database observed people (across population subgroups) spending approximately 5.5% of their time in vehicles, several studies have measured UFP concentrations in and immediately outside vehicles to estimate infiltration. Hudda et al. (2012) observed that I/O was positively associated with increasing AER for vehicles tested in Los Angeles, CA and Sydney, Australia each with recirculating air and outside air intakes. I/O increased with increasing vehicle speed and age, with a maximum of approximately 0.75 under recirculating conditions and of approximately 0.9 under outside air intake. Bigazzi and Figliozzi (2012) estimated I/O when a vehicle in Portland, OR was operated with windows down, windows up with outside air intake, and windows up with recirculating air. Under those

conditions, I/O decreased from 0.85 to 0.53 to 0.1–0.17, respectively. Knibbs et al. (2010) tested I/O for five vehicles and four ventilation settings (outdoor air intake with lowest and second lowest fan speed, recirculation on with lowest fan speed, recirculation on with fan off). Older model vehicles (prior to 2000) had I/O of 0.89–1.04 for the outdoor air intake settings and 0.29–0.47 for the recirculation settings. Models built after 2000 had I/O of 0.66–1.04 for outdoor air intake settings and 0.08–0.68 for recirculation settings. Yamada et al. (2016) took measurements along four road segments and inside one tunnel in the greater Tokyo, Japan area for particles smaller than and larger than 50 nm and using open air or recirculating air. When fresh air entered the vehicle, I/O ranged from 0.5 to 0.6 for particles smaller than 50 nm and from 0.8 to 0.9 for particles larger than 50 nm. When the test automobile's ventilation was operated in recirculation mode, infiltration ranged from 0.1 to 0.2 for particles smaller than 50 nm and from 0.2 to 0.9 for particles larger than 50 nm. In a tunnel in the greater Salzburg, Austria area, Madl et al. (2015) measured vehicle ventilation filtration efficiency for UFP, which can be used to interpret I/O by subtracting reported filtration efficiency from 1. They observed I/O of approximately 0.3 when the vehicle's standard ventilation setting was used, which reduced to 0.1 when the vehicle was put into recirculation mode. In all, these studies show that large variability in I/O occurs with both outdoor air intake and recirculation settings, but I/O tends to be higher for outdoor air intake.

Exposure to PM, particularly UFP, has been found to be elevated during bicycling and walking near roadways (Buonanno et al., 2013b; Hudda et al., 2012; Berghmans et al., 2009; Boogaard et al., 2009; Briggs et al., 2008). A study in Minneapolis, MN used city-wide traffic flows and a LUR model for particulate matter (including NC, BC mass, and $PM_{2.5}$) to analyze the relationship between bicycling or walking and PM exposure concentrations in different parts of the city (Hankey et al., 2017). The authors found that areas classified as high activity and high exposure made up approximately one-tenth of the total grid cells, but accounted for 20–44% of active travel.

Updated time-activity data and tools for assessing time-activity data have improved the general understanding of time-activity data and related uncertainties in recent years. Children were surprisingly found to spend less time outdoors than adults, but White respondents did spend more time outdoors than their Asian, Black, and Hispanic counterparts. New technologies to assess study participant location, errors related to study participant recall, and residential mobility have been used to determine that location-based errors are within 6% for short-term and long-term exposure assessment, while omission of residential mobility can result in a bias in the exposure estimate, resulting in biasing the health effect estimate for a study of long-term $PM_{2.5}$ exposure.

### 3.4.2.2 Spatial Variability in Concentrations

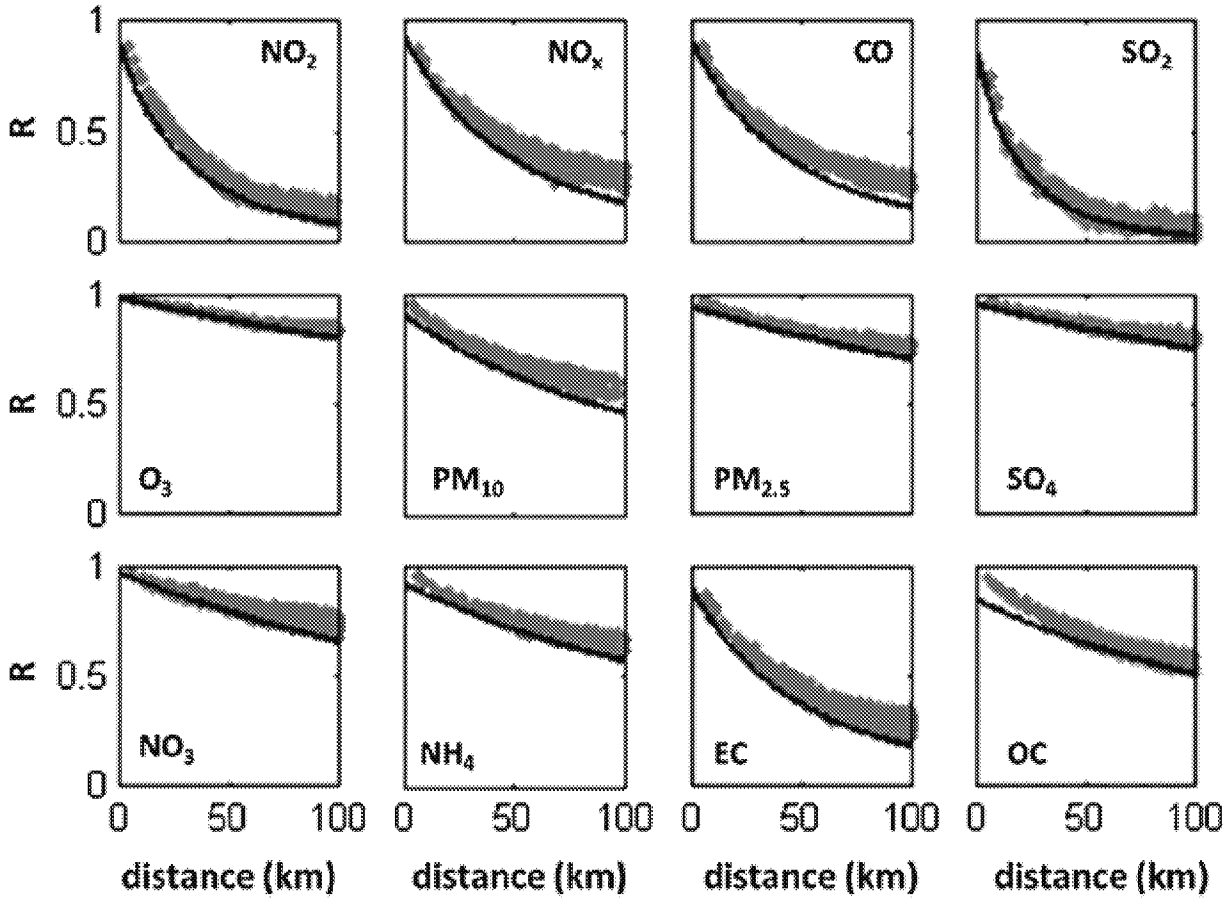
The 2009 PM ISA (U.S. EPA, 2009b) examined spatial relationships among $PM_{2.5}$ between AQS monitoring locations across neighborhood and urban scales. In general, this analysis suggested that correlations between monitors across space depended on the specific city's meteorology, topography, and

source mixture. For all cities studied, the between-monitor spatial correlations decreased with increasing distance between monitors. However, the correlation for $PM_{2.5}$ between Boston, MA monitor pairs was roughly Pearson $R = 0.8$ even when the monitors were 100 km apart. In contrast, correlation between $PM_{2.5}$ for Los Angeles monitor pairs was roughly Pearson $R = 0.2$ when the monitors were 100 km apart. The mountains and inversion patterns were thought to play a role in this comparatively low correlation. The 2009 PM ISA also investigated neighborhood scale monitor pair correlations among FRMs or FEMs in 15 CSAs or CBSAs and found that within 4 km, average correlation of Pearson $R = 0.93$ was maintained for a 4 km distance. At the time of the 2009 PM ISA, data were not available to study spatial variability in the concentration surface for $PM_{10-2.5}$ or UFP. Spatial distribution data for both UFP and $PM_{10-2.5}$ are still limited, especially for UFP. Data for UFP were available for two cities (Los Angeles, CA and Rochester, NY), and data from the Los Angeles study suggested that UFP had moderate spatial variability (coefficient of divergence [COD] between 0.2 and 0.6). It was thought that some background UFP reduced spatial variability, especially for particles larger than 40 nm (Section 2.5.1.2.4). Although some $PM_{10-2.5}$ data are available across the nation, micro-to-neighborhood scale data are not widely available at this size cut (Section 2.5.1.2.3). In cities where $PM_{10-2.5}$ measurements have been made in multiple locations, inter-monitor correlations were low. These limitations create uncertainty in characterizing spatial variability of exposure concentrations and its impact on interpreting results from epidemiologic studies, especially for long-term exposure to $PM_{10-2.5}$ and UFP.

Limitations in the use of ambient monitoring data to estimate exposure concentration arise when there is a lack of homogeneity and spatial autocorrelation of PM mass concentrations, which may occur for some size fractions and components (Baxter et al., 2013), causing the spatial range over which such estimates are used to vary widely. $PM_{10-2.5}$ and UFP concentration data tend to be more heterogeneous in space and hence more susceptible to spatial error (Section 2.5; Section 3.4.2.2). For large metropolitan areas, population exposure to primary anthropogenic components of PM (of any size fraction) may be substantially overestimated in terms of average concentration and temporal variation by the use of a fixed-site ambient monitor in close proximity to an industrial or energy generation source (Sarnat et al., 2015; Bell et al., 2011b). For example, traffic-related UFP and $PM_{2.5}$ components such as EC have elevated concentrations in close proximity to busy roadways (Zhu et al., 2009), potentially resulting in exposure misclassification (Ozkaynak et al., 2013; Bravo et al., 2012). Saturation sampling over longer time-scales may be used to ascertain spatial variation across an urban area, but at the expense of temporal resolution (Matte et al., 2013). Another limitation of using fixed-site ambient monitors to estimate exposure concentration is that ambient monitoring data can be incomplete due to missing data and sampling frequency limitations. Often missing data can be estimated using data from nearby monitors (e.g., by linear regression) or by temporal interpolation. Temporal interpolation can also be used for data analysis when the data are sampled with 1-in-3 or 1-in-6-day sampling frequencies (Junger and de Leon, 2015; Gomez-Carracedo et al., 2014; Junninen et al., 2004; Hopke et al., 2001), which is common for PM components. Interpolation schemes are used to capture hour-of-day and day-of-week trends. Estimates of mixing height using meteorological data and/or tracer component data are also used to improve the completeness of ambient monitor data.

Limited available $PM_{10-2.5}$ data for inter-site correlation and COD support previous statements that $PM_{10-2.5}$ tends to be spatially variable. Thornburg et al. (2009) measured correlation and COD in Detroit for personal multi-stage impactors measuring $PM_{10-2.5}$ and found Pearson $R = 0.28-0.63$ and COD = 0.17–0.41 during Summer and Pearson $R = 0.03-0.76$ and COD = 0.26–0.50 during Winter. Similarly, Lagudu et al. (2011) measured $PM_{10-2.5}$ using passive samplers and observed COD = 0.44–0.78 in the Spring and COD = 0.37–0.88 in the Fall. Neither the Thornburg et al. (2009) nor the Lagudu et al. (2011) studies included data for distances between specific monitors to ascertain if COD increased with increasing distance between samplers. This lack of data adds greater uncertainty to the characterization of $PM_{10-2.5}$ spatial variability.

Spatial variability of $PM_{2.5}$ components can vary among the components. Bell et al. (2011a) presented correlations for FRM or FEM pairs for seven $PM_{2.5}$ components ($NH_4^+$, EC, $NO_3^-$, OC, Si, $Na^+$, S) in a review paper. Bell et al. (2011a) observed that the bulk of the monitor-pair correlation is maintained relatively well (roughly Pearson $R = 0.8$) for $NH_4^+$, $NO_3^-$, and $SO_4^{2-}$ (Figure 3-6). Other components had wider variability in correlations even when the monitor pairs were closer together, as was the case for EC, Si, and $Na^+$. OC correlations were more variable than for $NH_4^+$, $NO_3^-$, or $SO_4^{2-}$ across monitor pair distances but not as variable as EC, Si, or $Na^+$. Dionisio et al. (2013) compared the coefficient of variation ($CV = \sigma/\mu$) of six air pollutants' concentrations across space using a hybrid AERMOD-background model of concentrations in the Atlanta, GA metropolitan area. They observed the following ordinal relationship of the covariates' median CVs: $NO_X$ (0.88) > CO (0.58) > EC (0.50) > $PM_{2.5}$ (0.13) > $O_3$ (0.07) > $SO_4$ (0.05) (see Figure 3-6). Likewise, Goldman et al. (2012) and Ivy et al. (2008) both used monitoring data from the Atlanta, GA metropolitan area to estimate spatial correlation functions, and they observed that the spatial correlograms for $O_3$, $PM_{10}$, $PM_{2.5}$, and the $PM_{2.5}$ components $SO_4^{2-}$, $NO_3^-$, $NH_4^+$, and OC were much less steep than for $NO_2$, $NO_X$, CO, $SO_2$, and EC. Hence, $PM_{2.5}$ was observed to be less spatially variable than copollutants frequently associated with traffic ($NO_X$, CO, EC) or industry ($SO_2$). Similarly, Goldman et al. (2012), Ivy et al. (2008) and Sajani et al. (2010) all observed less spatial variability of $PM_{10}$ compared with $NO_2$ or $NO_X$. If $PM_{10}$ were comprised primarily of $PM_{2.5}$, then these findings would be consistent with the Dionisio et al. (2013) results as well. These findings could reflect the influence of local sources and suggest that spatial variability of $PM_{2.5}$ components could have a large influence on monitor pair correlations for $PM_{2.5}$, with components with greater variation being influenced more by primary sources than components produced through secondary atmospheric chemistry.




Source: Permission pending Goldman et al. (2012).

**Figure 3-6 Spatial correlation of $PM_{2.5}$ components for monitor pairs described in the review study.**

It was known at the time of the 2009 PM ISA (U.S. EPA, 2009b) that spatial variability of $PM_{2.5}$ was lower than for $PM_{10-2.5}$ and UFP. Data to characterize $PM_{10-2.5}$ and UFP spatial concentration surfaces remain limited but generally support that comparison. More recent data for $PM_{2.5}$ components shows that components that are influenced by primary sources tend to be more spatially variable than components produced via atmospheric chemistry.

### 3.4.2.3 Instrument Accuracy and Precision

The influence of instrument error on health effect estimates from epidemiologic studies varies with study design. Inter-monitor comparison is often used to estimate instrument precision. Accuracy and

precision of ambient monitors is described in Section 2.5.4, and accuracy and precision for personal $PM_{2.5}$ monitors were described in the 2009 PM ISA (U.S. EPA, 2009b) and have not changed markedly since the last review.

More attention is given at present to $PM_{10-2.5}$, because those measurements were not as prevalent at the time of the 2009 PM ISA (U.S. EPA, 2009b). Errors associated with measurements of $PM_{10-2.5}$ are described in Section 2.4.2. Use of subtraction methods for estimating $PM_{10-2.5}$ concentration can lead to substantial errors. This is particularly true when the $PM_{10-2.5}$ is semivolatile. Clements et al. (2013) tested different methods for measuring $PM_{10}$ and $PM_{2.5}$ and calculating $PM_{10-2.5}$ via subtraction methods and found that the nonvolatile PM endemic to Colorado were measured with less error by instruments that did not account for semivolatile losses. Biases in calculated $PM_{10-2.5}$ concentrations caused reductions in correlation coefficients across sites, leading to an incorrect picture of spatial variability in $PM_{10-2.5}$ concentration across the test area.

A number of studies have characterized errors associated with measuring UFP (Section 2.4.3). UFP concentrations are often referred to without specific reference to size distribution. Some studies report number count as UFP, while other studies use mobility methods to impose an upper particle size limit of 100 nm or 250 nm. CPCs typically have lower size detection limits of 10 nm (Liu and Kim, 1977), while mobility have lower size detection limits of 1 nm (Kangasluoma et al., 2015; Lehtipalo et al., 2014; Kuang et al., 2012; Jiang et al., 2011; Vanhanen et al., 2011; Iida et al., 2008). Hence, use of CPCs in an epidemiologic study of short or long-term exposure may lead to an underestimation of the UFP exposure concentration.

For epidemiologic studies of short-term exposure, Goldman et al. (2010) investigated instrument precision error at locations where ambient monitors were collocated. Correlations between collocated measurements of $PM_{2.5}$ mass and components ($SO_4^{2-}$, $NO_3^-$, $NH_4^+$, EC, OC) ranged from Pearson $R = 0.85$ for OC to Pearson $R = 0.97$ for $PM_{2.5}$ mass. Depending on specific conditions such as sampler type (e.g., passive vs. continuous), meteorological conditions, or presence of semivolatile PM, instrument errors may vary in total magnitude or direction so that error is not always positively correlated with concentration. Analysis of instrument error compared with measured and true (i.e., simulated) concentrations for the Goldman et al. (2010) study suggested that the error was not correlated with either measured or true concentrations. Hence, the instrument error was neither pure Berkson error nor pure classical error, but it probably retained Berkson-like and classical-like characteristics. If instrument error and concentration are positively correlated, then error in the exposure concentration estimates will be larger in locations where there are more prevalent or stronger primary sources or at times when PM emissions are higher for a given location. Moreover, if error is positively correlated with concentration, then it would be anticipated that the magnitude of the instrument error is largest at times of day when emissions are highest.

Instrumentation bias could be anticipated to influence exposure concentration estimates used in long-term PM exposure studies in some situations. For example, geostatistical or LUR models may

underestimate exposure concentration when the model is fit using data from samples that have experienced negative artifacts due to volatility. Ambient temperature and relative humidity would not be expected to vary greatly within a city. Because climate and ambient sources are more likely to differ among cities, instrumentation error occurring when warm temperatures exacerbate evaporation could have a larger influence on the comparison of exposure concentrations among cities.

### 3.4.2.4 Model Accuracy and Precision

Error in PM exposure model predictions leads to some error in the health effect estimates from epidemiologic studies in which they are used. However, the implications of the type of errors depends upon the application. In statistical models used in epidemiologic studies, spatial, temporal, or concentration biases and errors may align with the health data being used, leading to potential errors and increased uncertainties in the health effect estimates (NRC, 2007).

The performance of the exposure models in recreating exposure estimates can impact the ensuing health analyses. LOOCV is often used to assess the exposure concentration estimates (Section 3.3.2), particularly for LUR. One issue with LOOCV is that monitoring sites can be clustered, such that removing a monitor that is near other monitors does not "stress" the model, because the value from the nearby monitors will lead to an accurate replacement value. That issue, along with the majority of sites being clustered in urban areas, can lead to seemingly good performance metrics that are not indicative of how well the method can estimate exposure concentrations away from monitoring sites. Given that exposure models are developed, in part, to estimate levels away from observation locations it is informative to have approaches to evaluate how well the method can estimate exposures in such cases. One approach that has been developed is to remove multiple monitors that are spatially grouped such that they are not being influenced by nearby observations (Lv et al., 2016). A related issue arises in LUR modeling. If a hold-out technique uses 90% of the data to both build and train the model, a different set of independent variables may be chosen than those in the full model. Wang et al. (2014) argued that a preferable approach is to build the full model and retrain it with 90% of the data. Wang et al. (2015) found that the LUR model performance ($R^2$ ranged from about 0.3 to 0.9 for $PM_{2.5}$) was positively associated with the magnitude of the health effect estimate. Alexeeff et al. (2015) conducted a simulation study using high resolution fields developed from MAIAC satellite data as the "true" field, and developed simulated spatiotemporal fields by kriging and using LUR. $R^2$ of the kriging and LUR methods ranged from about 0.24 to 0.98. They linked poor performance (e.g., lower $R^2$) with bias in the health effect estimates. Goldman et al. (2011) and Goldman et al. (2010) also found in a simulation study that increased exposure measurement error led to negative bias in the health outcomes and increased uncertainty. These, and related studies, show the potential impact of the accuracy of the exposure concentration metrics on bias and uncertainty in the health effect estimates in an epidemiologic study.

A major issue in using concentration surfaces estimated by CTMs for epidemiologic analyses is that the errors in the model inputs [e.g., emissions, (Koo et al., 2015; Xu et al., 2015; Hao and Larkin, 2014; Larkin et al., 2014; Paulot et al., 2014; Urbanski et al., 2011; Zhang et al., 2010b), meteorology (Digar et al., 2011), and surface characteristics] and parameters (e.g., chemical reaction, thermodynamic, and turbulence descriptions) lead to output errors, including time- or location-varying biases (Hogrefe et al., 2015; Koo et al., 2015; Porter et al., 2015; Hogrefe et al., 2014; Rao et al., 2014; Appel et al., 2013; Appel et al., 2012; Simon et al., 2012; Napelenok et al., 2011; Civerolo et al., 2010; Foley et al., 2010; Zhang et al., 2010b; Swall and Foley, 2009). Meteorological models, which are typically used to provide inputs to air quality models, have similar issues with inputs and parameters, thus leading to uncertain output fields that also have errors and uncertainties. Arrandale et al. (2011) also noted that mean bias and correlation varied by region with distinct spatial patterns. Given the potential for such errors, understanding how well such models can reproduce PM (including size and components) concentration fields for exposure or exposure concentration modeling is important.

Errors can be large, particularly when considering individual PM components (e.g., OC) or size fractions (e.g., UFPs) (Koo et al., 2015; Stanier et al., 2014; Zhang et al., 2010b). In terms of model parameters, this is often due to a fundamental lack of understanding of the processes, for example knowledge of the chemical reactions and products involving organic compounds or nucleation (Donahue et al., 2013; Shiraiwa et al., 2013; Worton et al., 2013; Chen et al., 2011; Donahue et al., 2011; Hoyle et al., 2011; Pierce et al., 2011; Zhang et al., 2010a; Kulmala et al., 2009; Nieminen et al., 2009; Kroll and Seinfeld, 2008; Kuang et al., 2008; Kulmala and Kerminen, 2008). Koo et al. (2015) conducted an extensive evaluation of two CTMs (CMAQ and CAMx) for the same domain, and found that the models, overall, performed similarly for $PM_{2.5}$, but differences were found upon further investigation (e.g., performance for individual PM components, and how the errors varied based on region and time). The Koo et al. (2015) study demonstrated that the same model will perform differently, sometimes dramatically, depending upon domain and time period such that performance in one application is not definitive support that performance will be similar in a different application. The limited availability of sub-24-hour PM mass concentration and component data has inhibited the evaluation of CTMs for simulating the diurnal variation of PM. Koo et al. (2015) used diurnally varying $PM_{2.5}$ compositional information available from SEARCH (Hansen et al., 2006; Hansen et al., 2003) to further assess CMAQ and CAMx model performance and found that, in addition to a low bias in OC and ammonium, during the summer the models also simulated a drop during the daytime that was not found in the observations. This additional bias could impact studies that used temporally finer-scale $PM_{2.5}$ exposure concentration estimates.

Due to the various potential errors in using air quality models to develop exposure concentration fields, Marmur et al. (2006b) and Marmur et al. (2006a) concluded that the direct use of CTMs in epidemiologic studies of acute health endpoints would lead to attenuation in the observed outcomes. Spatially- and temporally-varying biases and errors would also lead to questions of their use in epidemiologic studies of long-term exposures as well if the fields are not modified (Bravo et al., 2012),

such as by blending with PM concentrations derived from satellite observations, as discussed in Section 3.3.3.

### 3.4.3 Costressor Relationships

To assess the independent effects of PM in an epidemiologic study of health effects, it is necessary to identify (Bateson et al., 2007): (1) which copollutants (e.g., $NO_2$, CO, BC) and additional exposures (e.g., noise, traffic levels) are potential confounders of the health effect-PM relationship so that their correlation with PM can be tested and, if needed, accounted for in the statistical model; (2) the time period over which correlations might exist so that potential confounders are considered appropriately for the time period relevant for the epidemiologic study design (e.g., pollutants or other factors that are correlated over the long term might not be important for a short-term exposure epidemiologic study); and (3) the spatial correlation structure across multiple pollutants, if the epidemiologic study design is for long-term exposure. Given that a covariate must be correlated with both the exposure and the health effect to be a confounder, the potential for confounding of PM-related health effects can vary by the health endpoint of interest.

For copollutants that do show high correlations, copollutant models may be appropriate to adjust the effect estimate for each pollutant for the potential confounding effects of another pollutant if each pollutant is associated with the health effect (Tolbert et al., 2007). If one copollutant is a surrogate for an etiologically linked pollutant, copollutant models may attribute the effect to the copollutant measured with less error, regardless of whether it is the etiologically linked pollutant. In copollutant models where PM is measured with more error than a copollutant, a differential effect occurs where the health effect estimate of PM exposure may be lower than the health effect estimate of the copollutant, even if PM is the true causal agent (Zeger et al., 2000), as discussed in the 2009 PM ISA (U.S. EPA, 2009b). If this occurs, the health effect related to PM exposure would be underestimated or potentially not detected. Positive correlation between PM and the copollutant and between the exposure measurement errors of PM and the copollutant can add more negative bias to the PM health effect estimate. Spatial variability of concentration differs among the particle size spectrum, and this may cause more exposure measurement error in $PM_{10-2.5}$ or UFP compared with $PM_{2.5}$ (Section 3.4.2.2). Hence, if $PM_{2.5}$ is measured with less error than copollutants, it is likely that the effect will be attributed to $PM_{2.5}$.

This section considers temporal copollutant correlations and how relationships among copollutants may change in space. Temporal copollutant correlations are computed from the time series of copollutant concentrations for two different collocated monitors. Temporal correlations are informative for epidemiologic studies of short-term PM exposure when the sampling interval is less than a month for each of the copollutants. Temporal correlations are informative for epidemiologic studies of long-term PM exposures when sampling intervals are months-to-years. Spatial relationships are evaluated by comparing within-pollutant variation across space for different pollutants. The following sections review

coexposures that can potentially confound the relationship between a health effect and PM exposure over different temporal and spatial resolutions.

### 3.4.3.1 Temporal Relationships among Ambient PM and Copollutant Exposures

AQS data presented in the 2009 PM ISA (U.S. EPA, 2009b) demonstrated most correlations between $PM_{2.5}$ and gaseous copollutants were typically between −0.2 and 0.8 with average and median values around 0.2 to 0.5. Correlations between $PM_{2.5}$ and $PM_{10-2.5}$ were observed in a similar range. Given limited data for $PM_{10-2.5}$ at the time when the 2009 PM ISA was written, correlations between $PM_{10-2.5}$ and gaseous copollutants were not presented.

To place the copollutant correlation discussion in the context of the epidemiologic studies, we present the correlation data for the epidemiologic studies in CHAPTER 5, CHAPTER 6, CHAPTER 7, CHAPTER 8, CHAPTER 9, CHAPTER 10, and CHAPTER 11 that reported correlations of $PM_{2.5}$, $PM_{10-2.5}$, or UFP with copollutants. Figure 3-7, Figure 3-10, and Figure 3-13 (for $PM_{2.5}$, $PM_{10-2.5}$, and UFP, respectively) plot study data for correlations with gaseous copollutants $O_3$, CO, $SO_2$, $NO_2$, and $NO_X$ and with particulate copollutants. More data were available for $PM_{2.5}$ compared with $PM_{10-2.5}$ or UFP (as NC, based on the assumption that the majority of particles are smaller than 100 nm), and so Figure 3-7 is divided into four panels for all data combined, acute timescales within 1 hour, short-term timescales between 1 hour and 2 weeks (with most data obtained at a 24-hour timescale), and long-term timescales longer than 2 weeks. Only 24-hour data were available for $PM_{10-2.5}$ and UFP correlation data.

For acute and short-term timescales (within 1 hour and 2 weeks, respectively), median correlations of $PM_{2.5}$ with copollutants were ordered $CO > NO_2 > SO_2 > NO_X > O_3$ (Figure 3-7). Acute data were relatively sparse but produced median correlations that were lower than those for short-term. Because data were combined across studies, Figure 3-7 includes both Pearson and Spearman correlations. Short-term correlations for CO and $NO_2$ reached as high as $R = 0.9$, while roughly 20% of the short-term correlations between $PM_{2.5}$ and $O_3$ were negative. Correlation data between UFP and $O_3$ were limited to one study (Kearney et al., 2011), and three of four reported correlations were negative in contrast to the mostly positive correlations between $PM_{2.5}$ and $O_3$ (Figure 3-13). Data for short-term correlations of $PM_{2.5}$ with $PM_{10-2.5}$ and UFP were around $R = 0.5$, although data were also sparse for these comparisons. Median correlations of $PM_{10-2.5}$ and gases ranged between $R = 0.3$ and $R = 0.5$, although limited data were available for these comparisons. Correlations of $PM_{10-2.5}$ with CO and $NO_2$ were around $R = 0.5$, potentially indicating some commonality of sources, such as traffic emissions of CO and (indirectly) of $NO_2$ with $PM_{10-2.5}$ generated by brake dust (Section 2.4.2). For short-term correlations of UFP with copollutant gases and particles, median correlations were 0.5 for $NO_2$ and lower for everything else. It is possible that low correlations could be related to the short lifetime of UFP relative to other PM size fractions. However, because limited data for UFP correlations were available, few conclusions can be

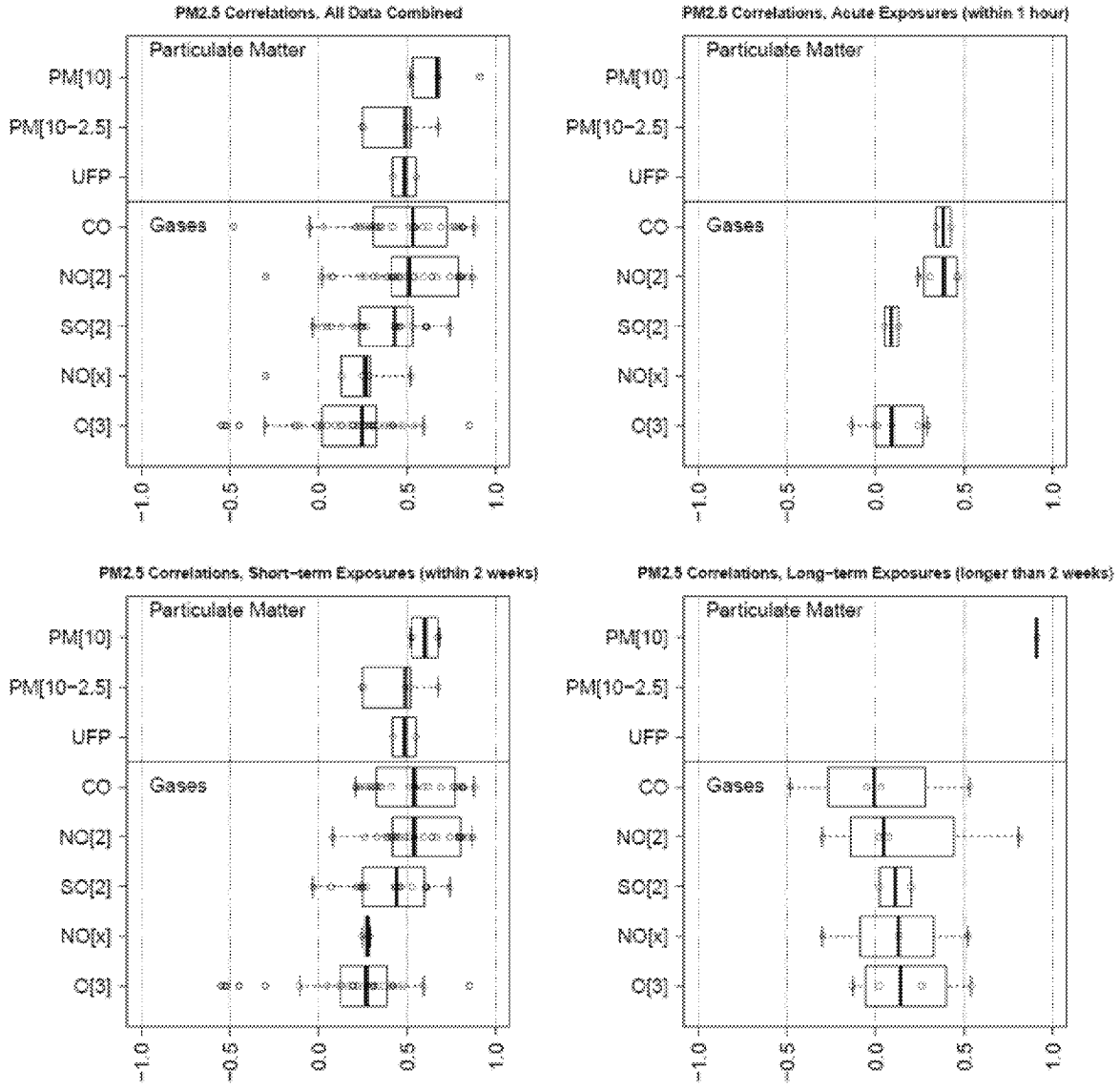
drawn. Because data were combined across studies, Figure 3-13 also includes both Pearson and Spearman correlations.

Median long-term correlations (i.e., longer than 2 weeks) between $PM_{2.5}$ and copollutants follow a pattern opposite to that for short-term correlations: $O_3 > NO_X > SO_2 > NO_2 > CO$ (Figure 3-7). Median correlations were between $R = 0$ and $R = 0.2$. Limited quantity of data existed for long-term correlations between $PM_{2.5}$ and copollutants and no data existed for long-term correlations of $PM_{2.5}$ with $PM_{10-2.5}$ or UFP. Moreover, overlapping 25th-to-75th percentile and 5th-to-95th percentile intervals reduce confidence in the comparison.

For comparison to the epidemiologic data, short-term (24-hour average) correlations of $PM_{2.5}$ and copollutants and of $PM_{10-2.5}$ and copollutants were studied using air quality data from collocated monitors reported within the U.S. EPA AQS repository system during 2013–2015. 438 sites met the 75% data completeness criteria presented in Section 2.5.1.1. Pearson correlations were used to evaluate temporal correlations among ambient $PM_{2.5}$ concentrations and NAAQS copollutant concentrations. Figure 3-8 displays the distribution of correlations between NAAQS copollutants and 24-hour $PM_{2.5}$ for annual data for 2013–2015, and Figure 3-9 displays the distribution of correlations broken down by season. For CO, $SO_2$, and $NO_2$, 1-hour daily max concentrations are used, while for $O_3$, 8-hour daily max concentrations are considered. Annual and seasonal copollutant correlation plots for 24-hour $PM_{10-2.5}$ are provided in Figure 3-11 and Figure 3-12.

Across seasons, 24-hour average $PM_{2.5}$ and $PM_{10-2.5}$ concentrations reported in the AQS consistently have the highest correlations with $PM_{10}$ concentrations (median Pearson $R = 0.7–0.8$ for $PM_{2.5}$, median Pearson $R = 0.7–0.9$ for $PM_{10-2.5}$) (Figure 3-9, Figure 3-12). This could occur if $PM_{2.5}$ were a large contributor to $PM_{10}$, if $PM_{2.5}$ and $PM_{10-2.5}$ were of the same source, or if $PM_{2.5}$ and $PM_{10-2.5}$ were of different sources whose emissions were coordinated in time. Correlations between $PM_{2.5}$ concentrations and $PM_{10-2.5}$ concentrations are lower than either size fraction's correlation with $PM_{10}$ across seasons (median Pearson $R = 0.2–0.5$), with lowest correlations in winter. This is consistent with observations from the epidemiology literature (Figure 3-7, Figure 3-10), although data for $PM_{10-2.5}$ correlations are limited. Figure 3-7 and Figure 3-10 do not distinguish between Pearson and Spearman correlations, because data are combined across studies. In the summer and spring, correlations of $PM_{2.5}$ with $SO_2$, $NO_2$, and CO are all roughly $R = 0.2$. In the fall and winter, however, correlations of $PM_{2.5}$ are ordered as $CO > NO_2 > SO_2$, consistent with correlations reported in the epidemiology literature (Figure 3-9). Higher correlations of CO and $NO_2$ with $PM_{2.5}$ may be indicative of combustion sources. Correlation of $PM_{2.5}$ and $O_3$ is highest during the summer (median Pearson $R \sim 0.45$) and is negative during the winter. High summer correlations could reflect photooxidation to produce simultaneously higher levels of $O_3$ and secondary PM (Section 2.3.2.3), (U.S. EPA, 2013). Median correlations of $PM_{10-2.5}$ with $SO_2$, $NO_2$, CO, and $O_3$ were all in the range of $R = 0.1–0.3$ across seasons. This may reflect the origin of $PM_{10-2.5}$ largely as dust rather than by combustion, other industrial processes, or photochemistry.

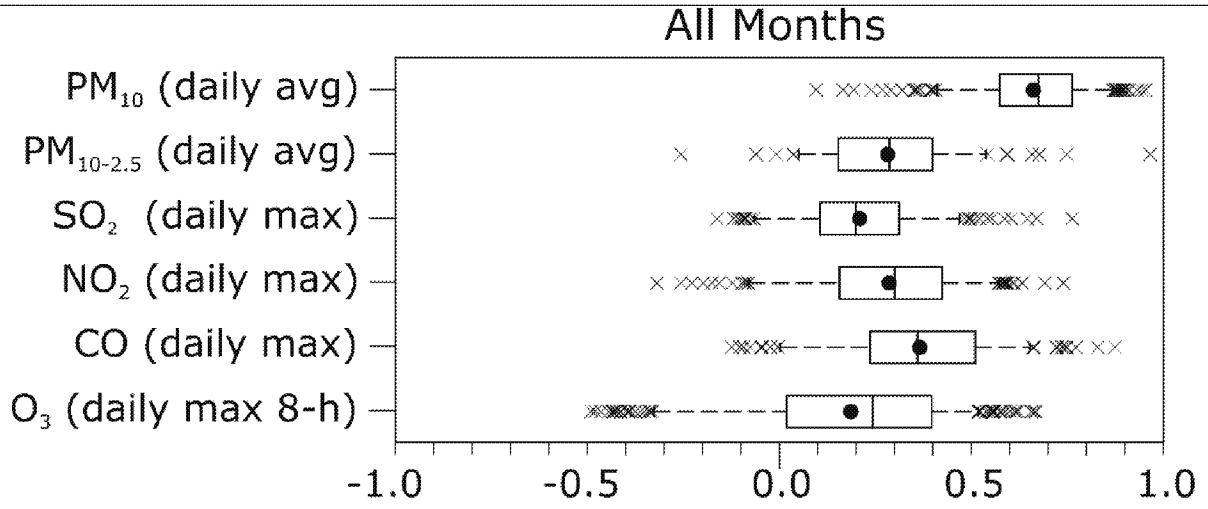
Correlation data from epidemiology studies (Figure 3-10) are higher for CO and $NO_2$, but only a limited number of studies reported those correlations.




Based on epidemiologic studies reporting correlations in CHAPTER 5, CHAPTER 6, CHAPTER 7, CHAPTER 8, CHAPTER 9, CHAPTER 10, and CHAPTER 11.

Source: Permission pending, References listed in Richmond-Bryant (2018).

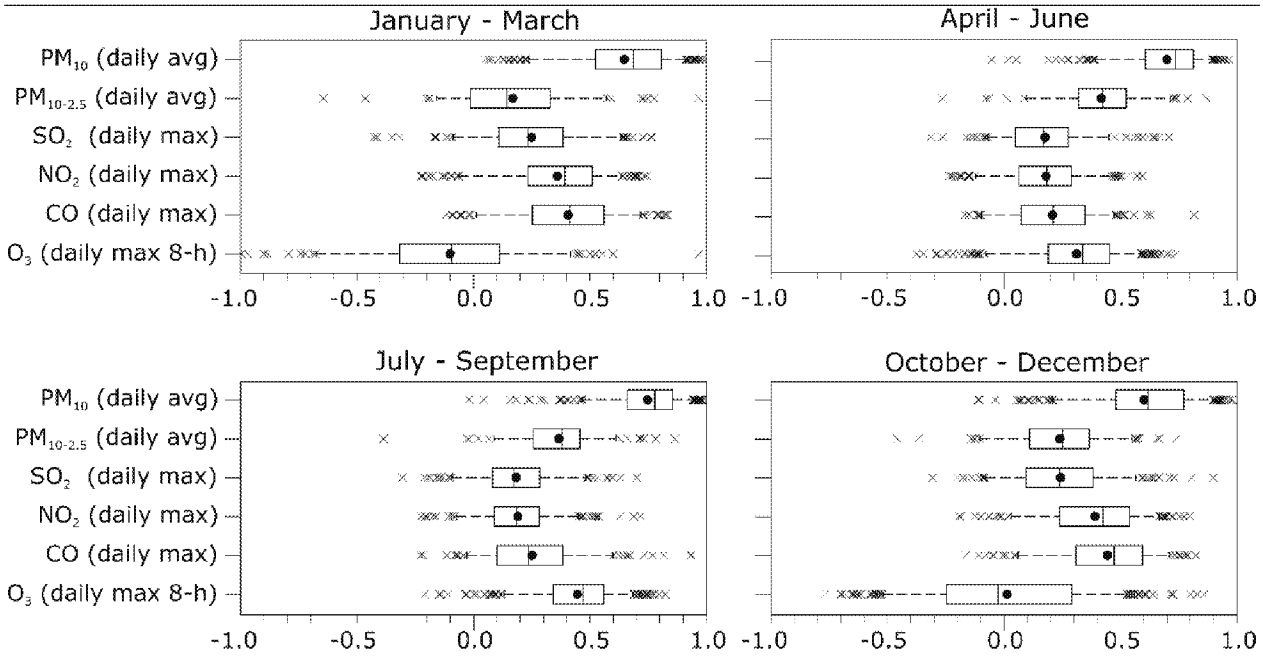
**Figure 3-7 Correlations between $PM_{2.5}$ and copollutants for all data combined (top left), timescales within 1 hour (top right), short-term timescales within 2 weeks (bottom left), and long-term timescales greater than 2 weeks (bottom right).**




CO = carbon monoxide; $NO_2$ = nitrogen dioxide; $O_3$ = ozone; $PM_{10-2.5}$ = particulate matter with a nominal aerodynamic diameter less than or equal to 10 µm and greater than 2.5 µm; $PM_{10}$ = particulate matter with a nominal aerodynamic diameter less than or equal to 10 µm; S = sulfur.

Note: Shown are the median (line), mean (circle), and inner-quartile range (box), 5th and 95th percentile (whiskers) and extremes (x's).

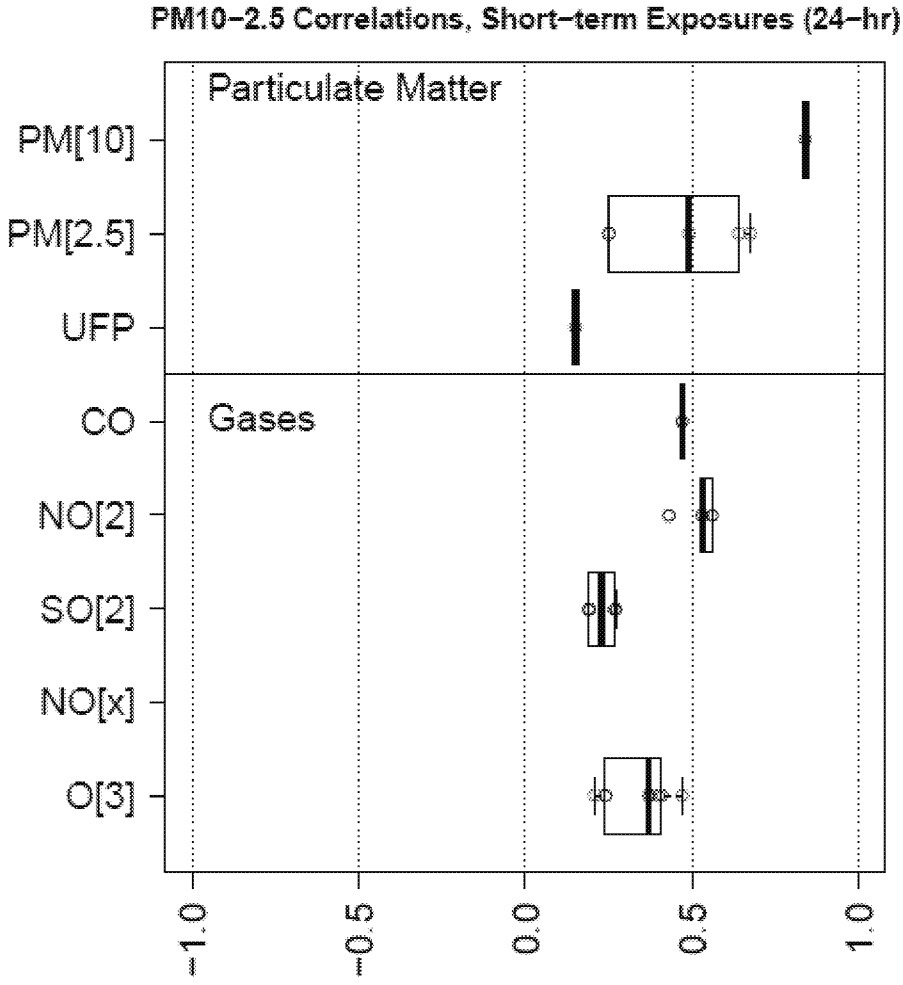
**Figure 3-8 Distribution of Pearson correlation coefficients for annual 24-hour average concentration of $PM_{2.5}$ with collocated copollutants from the Air Quality System during 2013−2015.**




CO = carbon monoxide; $NO_2$ = nitrogen dioxide; $O_3$ = ozone; $PM_{10-2.5}$ = particulate matter with a nominal aerodynamic diameter less than or equal to 10 µm and greater than 2.5 µm; $PM_{10}$ = particulate matter with a nominal aerodynamic diameter less than or equal to 10 µm; S = sulfur.

Note: Shown are the median (line), mean (circle), and inner-quartile range (box), 5th and 95th percentile (whiskers) and extremes (x's).

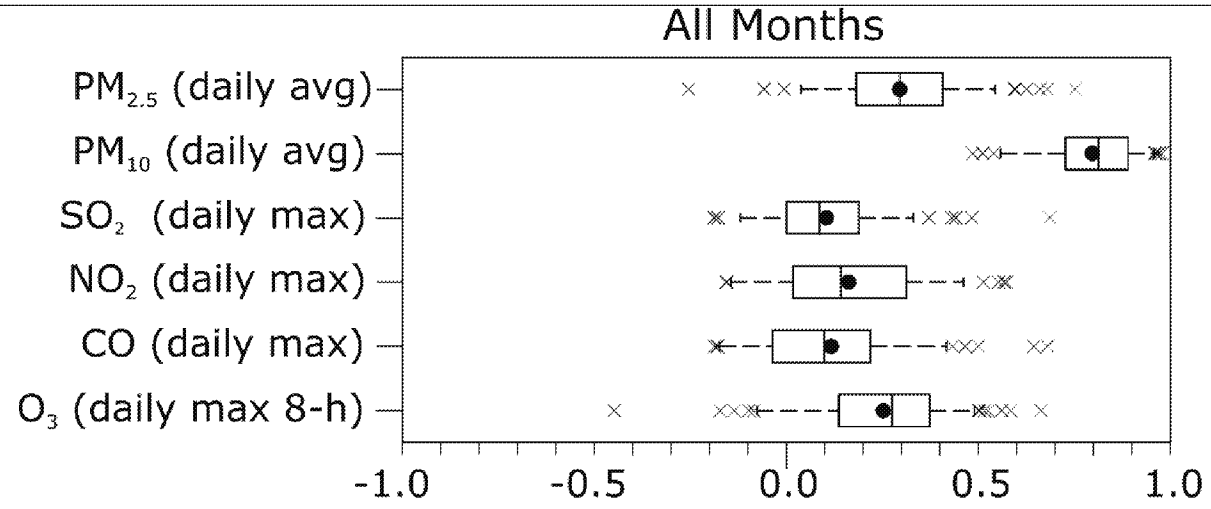
**Figure 3-9 Distribution of Pearson correlation coefficients for comparison of seasonal 24-hour average concentration $PM_{2.5}$ with collocated copollutants from the Air Quality System during 2013–2015.**




Note: Only 24-hour data were available for $PM_{10-2.5}$. Based on epidemiologic studies reporting correlations in CHAPTER 5, CHAPTER 6, CHAPTER 7, CHAPTER 8, CHAPTER 9, CHAPTER 10, and CHAPTER 11.

Source: Permission pending, (Chen et al. (2015); Cheng et al. (2015); Michikawa et al. (2015); Qiu et al. (2014); Raza et al. (2014); Alessandrini et al. (2013); Qiu et al. (2013); Rosenthal et al. (2013); Wichmann et al. (2013); Qiu et al. (2012); Atkinson et al. (2010)).

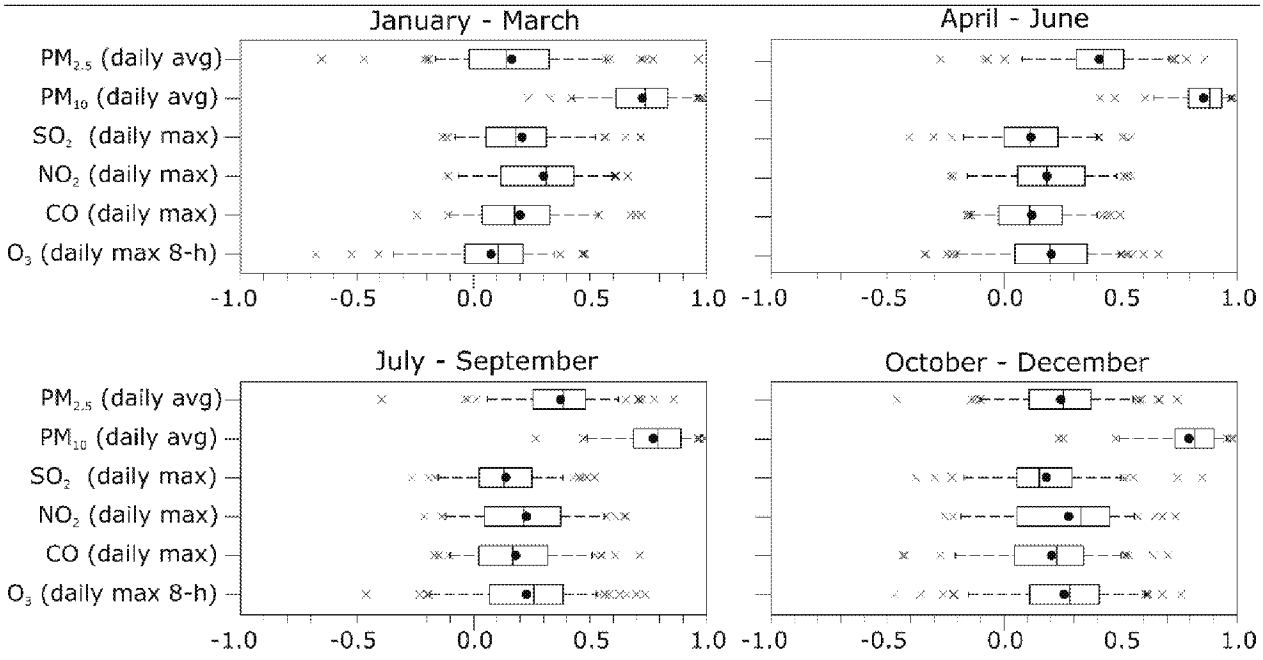
**Figure 3-10 Pearson correlations between $PM_{10-2.5}$ and copollutants for short-term exposures.**




CO = carbon monoxide; $NO_2$ = nitrogen dioxide; $O_3$ = ozone; $PM_{10-2.5}$ = particulate matter with a nominal aerodynamic diameter less than or equal to 10 µm and greater than 2.5 µm; $PM_{10}$ = particulate matter with a nominal aerodynamic diameter less than or equal to 10 µm; S = sulfur.

Note: Shown are the median (line), mean (circle), and inner-quartile range (box), 5th and 95th percentile (whiskers) and extremes (x's).

**Figure 3-11 Distribution of Pearson correlation coefficients for annual 24-hour average concentration of $PM_{10-2.5}$ with collocated copollutants from the Air Quality System during 2013−2015.**

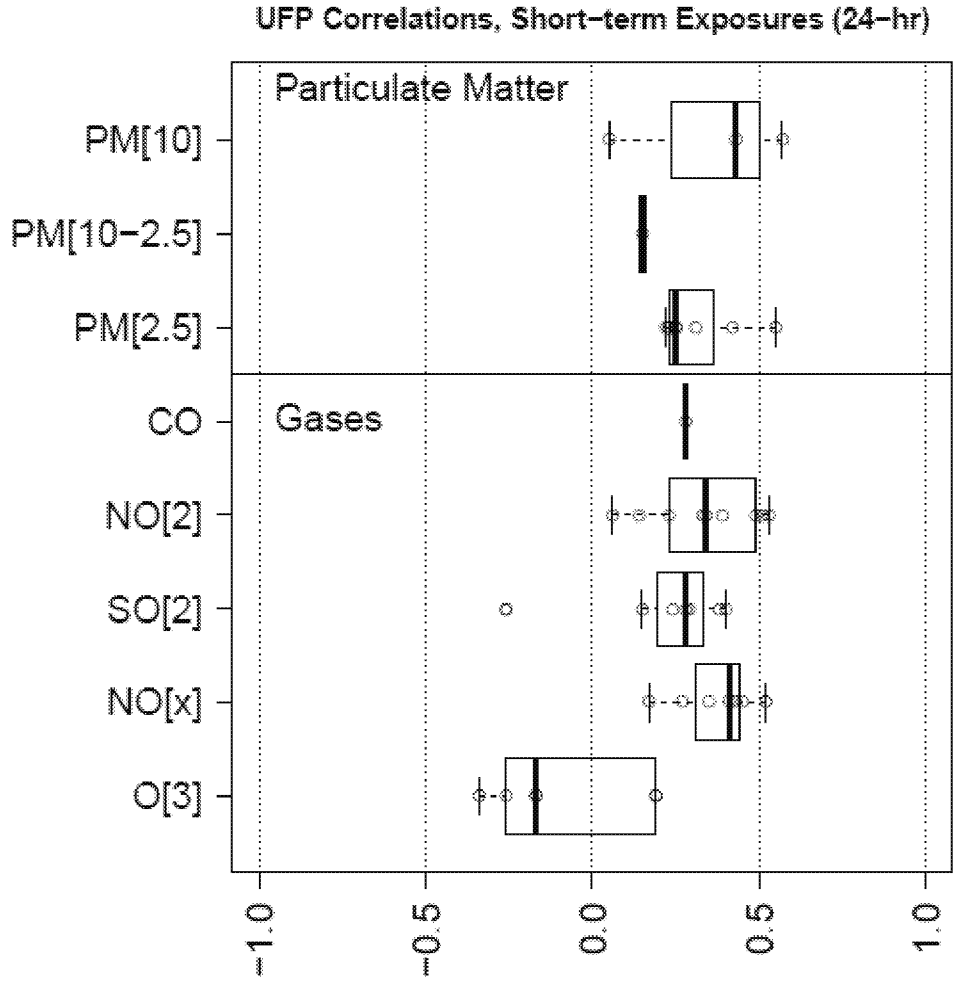



CO = carbon monoxide; $NO_2$ = nitrogen dioxide; $O_3$ = ozone; $PM_{10-2.5}$ = particulate matter with a nominal aerodynamic diameter less than or equal to 10 µm and greater than 2.5 µm; $PM_{10}$ = particulate matter with a nominal aerodynamic diameter less than or equal to 10 µm; S = sulfur.

Note: Shown are the median (line), mean (circle), and inner-quartile range (box), 5th and 95th percentile (whiskers) and extremes (x's).

**Figure 3-12 Distribution of Pearson correlation coefficients for comparison of seasonal 24-hour average concentration of $PM_{10-2.5}$ with collocated copollutants from the Air Quality System during 2013–2015.**

Limited data were available from the peer-reviewed literature for correlations of UFP concentration with concentrations of other PM size fractions or of gases (Figure 3-13). Median Pearson correlations around $R = 0.5$ were reported for UFP with $PM_{2.5}$ and with $NO_2$ and $NO_X$. Without more data to identify copollutant relationships for UFP, it is difficult to interpret these data.




Note: Only 24-hour data were available. Based on epidemiologic studies reporting correlations in Chapters 5−11.

Source: Permission pending, Iskandar et al. (2012); Kearney et al. (2011); Leitte et al. (2011); Andersen et al. (2010); Atkinson et al. (2010); Belleudi et al. (2010).

**Figure 3-13 Correlations between UFP and copollutants for short-term exposures.**

### 3.4.3.2 Spatial Relationships among Ambient PM and Copollutant Exposures

When an epidemiologic study design relies on spatial contrasts to draw conclusions, such as for an epidemiologic study of long-term exposure, unmeasured spatial correlation between copollutants may lead to positive bias in the health effect estimate for each of the pollutants included in the model. Paciorek (2010) performed simulations and analyzed case study data (of the relationship between birth weight data and BC concentrations in eastern Massachusetts) to test the effect of spatial errors on health effect estimates in long-term exposure epidemiologic studies. In this study, Paciorek (2010) selected BC as a

PM component because it is spatially variable. He identified unmeasured spatial confounding as a key driver in biasing health effect estimates in a spatial regression. Paciorek (2010) maintained that bias can be reduced when variation in the exposure concentration metric occurs at a smaller spatial scale than that of the unmeasured confounder. The findings of Paciorek (2010) would be expected to be more significant for more spatially-variable $PM_{10-2.5}$, UFP, and BC than for $PM_{2.5}$, for which less spatial error would be anticipated.

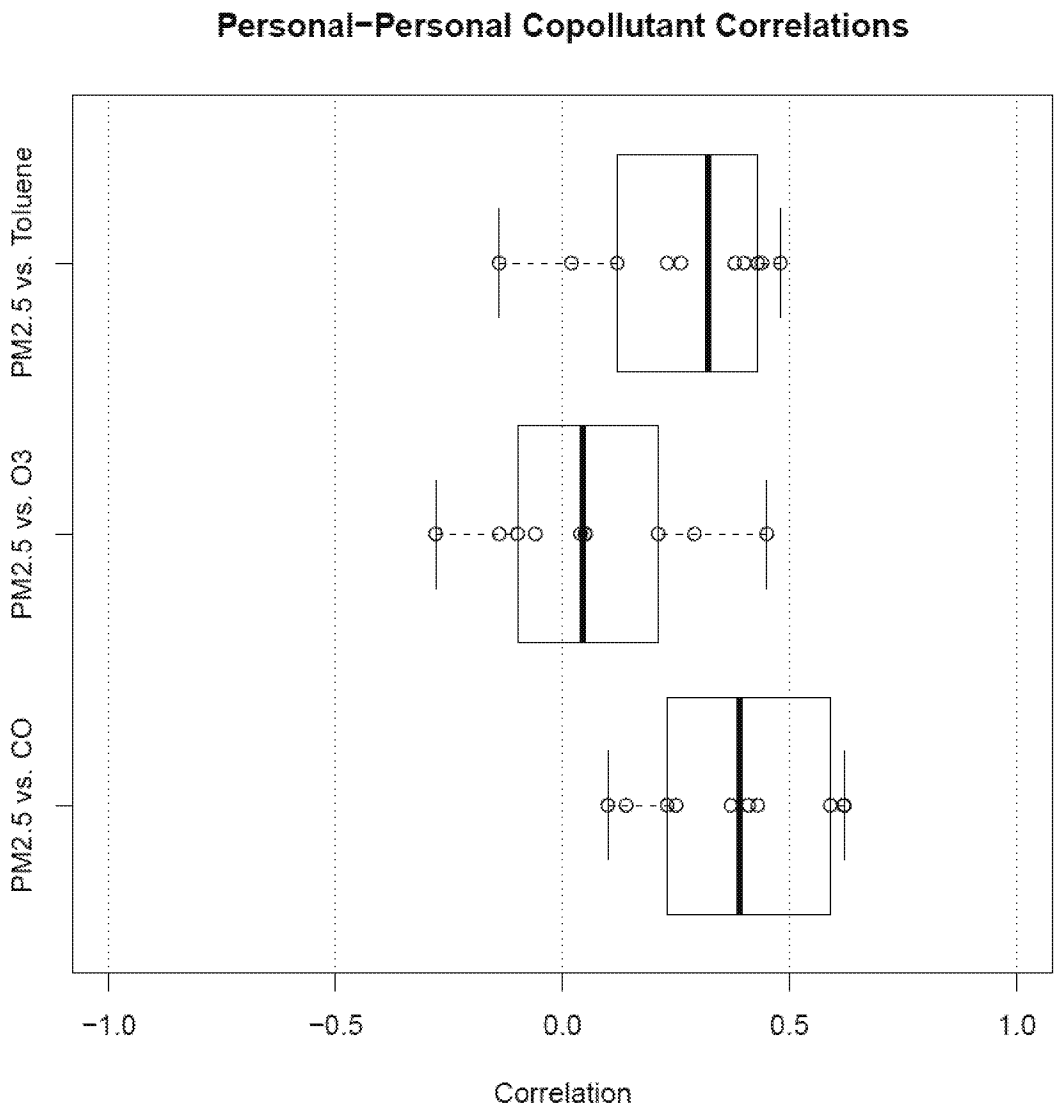
#### 3.4.3.3 Personal and Indoor Relationships between PM and Copollutant Exposures

No new studies on relationships among personal and ambient copollutants had been performed since the 2009 PM ISA (U.S. EPA, 2009b). Those data are presented graphically in Figure 3-14, Figure 3-15, and Figure 3-16. Figure 3-14 displays copollutant correlations among personal exposures to $PM_{2.5}$, toluene, $O_3$, and CO. The data from Chang et al. (2000) were obtained in Baltimore, MD in the summer of 1998 and winter of 1999. Median correlations were 0.39 for the personal-personal relationship for $PM_{2.5}$ versus CO, 0.32 for $PM_{2.5}$ versus toluene, and 0.045 for $PM_{2.5}$ versus $O_3$. Correlations were highest when personal measurements were obtained outdoors away from the road during the summer for $PM_{2.5}$ versus $O_3$ and $PM_{2.5}$ versus CO during the summer and for $PM_{2.5}$ versus toluene during the winter. The higher correlations obtained away from the road may reflect the secondary nature of much of the measured $PM_{2.5}$.

Median personal-ambient slopes between $PM_{2.5}$ and gaseous copollutants are generally between 0 and 0.5, as shown in Figure 3-15. These data were obtained from Koutrakis et al. (2005), Sarnat et al. (2005), Sarnat et al. (2001), and Sarnat et al. (2006b) from Boston, MA, Baltimore, MD, and Steubenville, OH. Median relationships of personal $PM_{2.5}$ exposure with ambient gaseous copollutant concentrations were higher with more variability than those of personal $SO_4^{2-}$ exposures with ambient gas concentrations, indicating that nonambient $PM_{2.5}$ exposure may have amplified these relationships and added uncertainty. Data were more limited for relationships between personal EC concentration and ambient gaseous copollutant concentrations, but these tended to be lower as well. Greater variability occurred in some cases for the relationships between personal exposure to gaseous copollutants and ambient concentrations of $PM_{2.5}$, EC, and $SO_4^{2-}$, perhaps as a result of limited amounts of data.

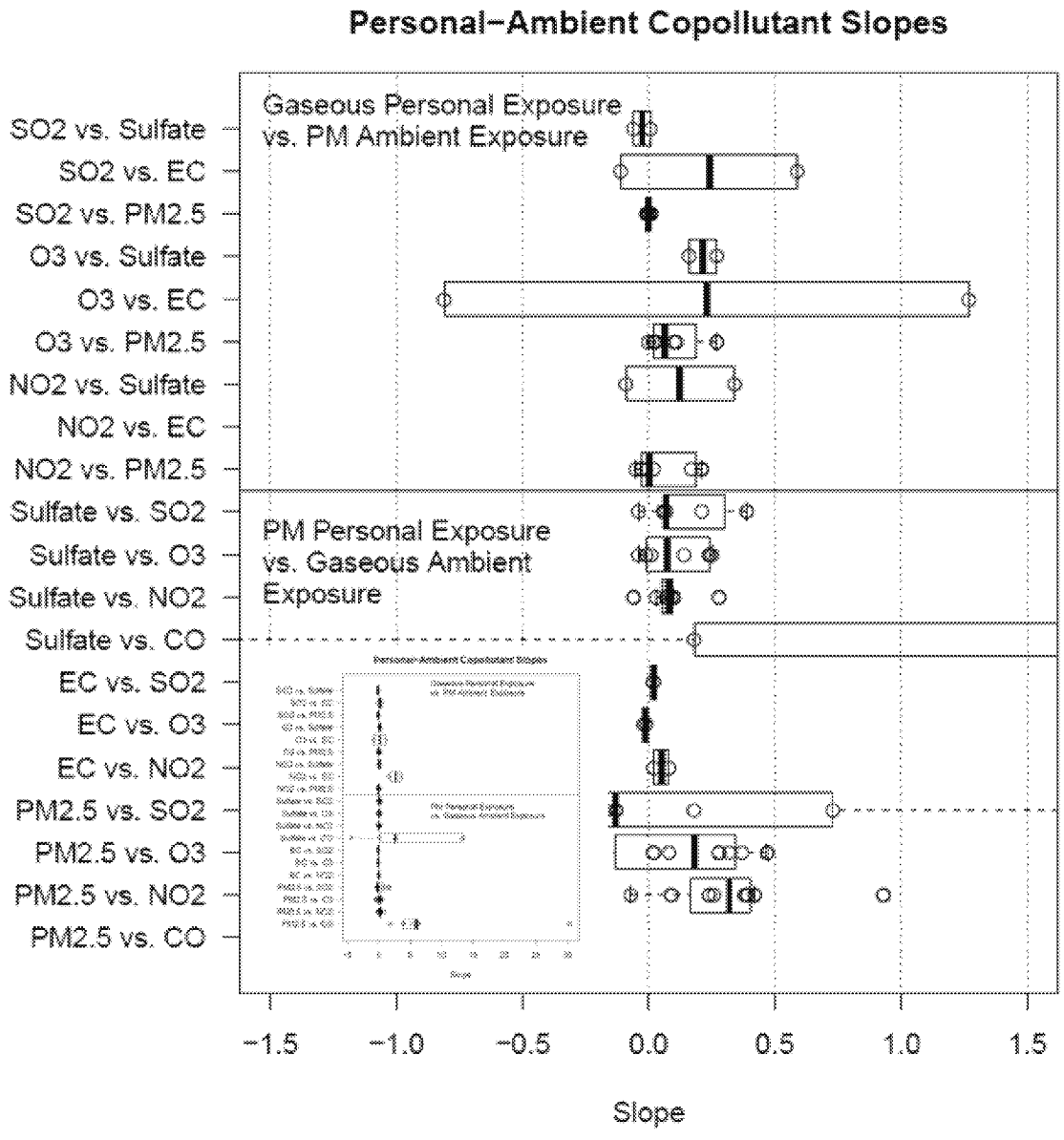
Median slopes for the relationship between personal exposure to PM or $SO_4^{2-}$ with gaseous copollutants ($NO_2$, $O_3$, and $SO_2$) tended to be between 0 and 0.5 (Figure 3-16). The exception was the relationship between $PM_{2.5}$ and $SO_2$, which was negative but of similar magnitude. These data were obtained from Koutrakis et al. (2005), Sarnat et al. (2005), and Sarnat et al. (2001). A slight reduction in median slope along with smaller data intervals were observed when personal $SO_4^{2-}$ exposure was used in lieu of personal $PM_{2.5}$ exposure, suggesting that the nonambient component of personal exposure may have influenced these relationships. Nonambient sources of $O_3$ and $SO_2$ are much less prevalent, so it is unlikely that they would have influenced their respective relationships. Although $NO_2$ does have indoor

(indirect) sources, variability in these relationships was lower than for the other gaseous copollutant exposures.



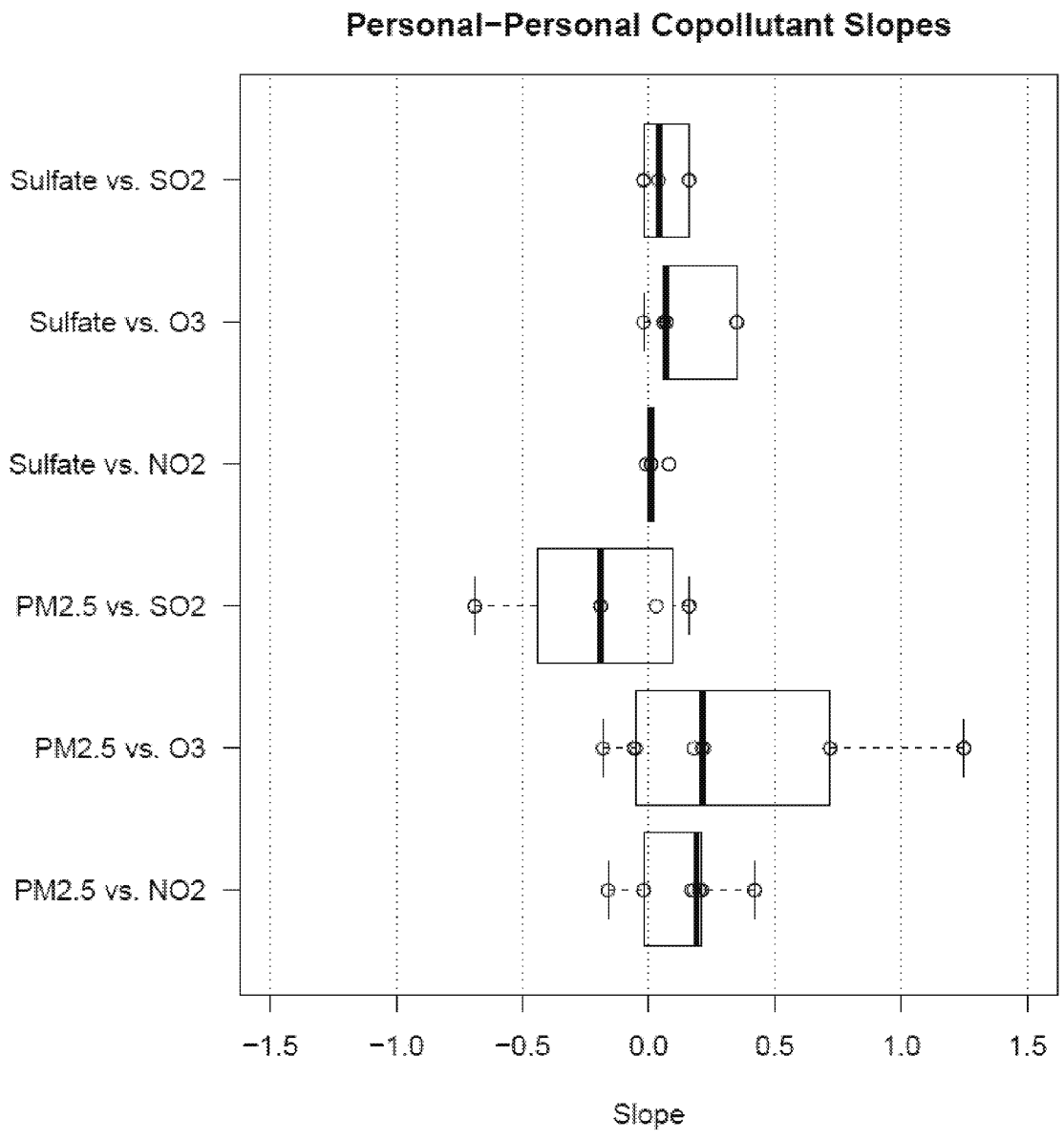

Source: Permission pending, (Chang et al., 2000).

**Figure 3-14 Correlations between personal exposure to $PM_{2.5}$ mass and personal exposure to gases.**




Note: Outliers for $NO_2$ vs. EC, $SO_4^{2-}$ vs. CO, and $PM_{2.5}$ vs. CO are shown on the small inset figure.
Source: Permission pending, Sarnat et al. (2006b); Koutrakis et al. (2005); Sarnat et al. (2005); Sarnat et al. (2001).

**Figure 3-15 Slopes for personal-ambient relationships. Top: Personal exposure to gaseous copollutants related to ambient exposure to $PM_{2.5}$ mass or EC or $SO_4^{2-}$ components.**




Source: Permission pending, Koutrakis et al. (2005); Sarnat et al. (2005); Sarnat et al. (2001).

**Figure 3-16 Slopes for personal-personal relationships between $PM_{2.5}$ mass or $SO_4^{2-}$ component and gaseous copollutants.**

### 3.4.3.4 Traffic-related Noise

The 2009 PM ISA (U.S. EPA, 2009b) did not consider the relationship of PM with traffic-related noise levels. Recent evidence is inconsistent regarding the correlations of PM concentrations with traffic and noise levels (HEI, 2010). There are differences among the studies exploring the health effects of PM and noise regarding size cut of PM measured, road type, and surrounding features. Hence, the role of traffic and noise as confounders or independent variables in the relationship between health effects and PM exposure is unclear.

Several studies have examined the relationship of traffic-related noise with PM concentrations. Kheirbek et al. (2014) added noise level meters to the dense New York, NY monitoring project described in Ross et al. (2013) and observed that 1-week average noise level (measured as dB[A]), obtained at 60 locations during Fall 2012, correlated with Pearson $R = 0.45$ for $PM_{2.5}$ concentration and Pearson $R = 0.62$ for BC concentration. Boogaard et al. (2009) measured UFP, $PM_{2.5}$, and noise (measured as dB[A]) while bicycling on scripted 10- to 20-minute routes for ten cities in The Netherlands and found a median correlation of Pearson $R = 0.34$ across cities for UFP and noise while the median correlation was Pearson $R = 0.009$ for $PM_{2.5}$ and noise. Gan et al. (2012b) calculated the correlations among air pollutants and noise from road traffic and aircraft using 5-minute data from 103 sites in Vancouver, BC, Canada during 2003 (dates not stated). They observed lower correlations for $PM_{2.5}$ concentration with road traffic noise (Spearman $R = 0.14$) compared with that for BC (Spearman $R = 0.45$). However, correlations between $PM_{2.5}$ and aircraft noise were higher (Spearman $R = 0.31$) than for BC (Spearman $R = -0.07$). Over a 5-year average, Gan et al. (2012a) reported the correlation between $PM_{2.5}$ concentration and noise from road traffic to be Spearman $R = 0.14$. Reported correlation of 5-year average BC concentration with BC concentration had a Spearman $R = 0.44$. These findings are consistent with the short-term observations reported in Gan et al. (2012b).

Ross et al. (2011) also examined relationships of different frequency noises with $PM_{2.5}$ and EC concentrations using continuous monitors collecting 48,000 samples per second for six 24-hour periods in August 2009. Ross et al. (2011) measured the relationships between traffic level, noise, and concentrations of $PM_{2.5}$ and EC in New York, NY as part of the Ross et al. (2013) study. Unweighted noise of all frequencies was uncorrelated with $PM_{2.5}$ concentration (Spearman $R = 0.20$) but correlation increased for EC concentration (Spearman $R = 0.35$) for all times. Correlations were higher for medium frequency noise ($PM_{2.5}$: Spearman $R = 0.20$; EC: Spearman $R = 0.39$) compared with high frequency noise ($PM_{2.5}$: Spearman $R = 0.14$; EC: Spearman $R = 0.15$) but were similar for low frequency noise ($PM_{2.5}$: Spearman $R = 0.19$; EC: Spearman $R = 0.32$). Correlations between $PM_{2.5}$ and low frequency noise (Spearman $R = 0.3$) were higher during rush hour than at night for low frequency noise or for any time for medium and high frequency noise. At night, high frequency noise had a higher correlation with EC concentration (Spearman $R = 0.4$).

Distance to road has also been observed to influence the relationship between noise and PM concentration as a surrogate for exposure concentration. The Gan et al. (2012b) study described above

also reported Spearman correlations between 5-minute average. A-weighted equivalent noise (i.e., noise level that is adjusted to noise perception by the human ear) and concentrations of $PM_{2.5}$ and BC for buffers of 50 m and 150 m of a highway (defined as A1 and A2 roads) and a major road (defined as A1, A2, and A3 roads). Correlations for $PM_{2.5}$ and noise were Spearman $R = 0.02$ within 50 m of the highway, Spearman $R = 0.03$ within 150 m, and Spearman $R = 0.17$ when further than 150 m. For a major road, correlations for $PM_{2.5}$ and noise were Spearman $R = 0.24$ within 50 m, Spearman $R = 0.15$ within 150 m, and Spearman $R = 0.14$ when further than 150 m. Results for correlations between BC and noise were higher than for correlations between $PM_{2.5}$ and noise, and they were more consistent between highways (within 50 m: Spearman $R = 0.17$, within 150 m: Spearman $R = 0.38$, further than 150 m: Spearman $R = 0.41$) and major roads (within 50 m: Spearman $R = 0.26$, within 150 m: Spearman $R = 0.46$, further than 150 m: Spearman $R = 0.31$). Allen et al. (2009) studied the relationship between UFP concentration, and 5-minute average A-weighted equivalent noise for 105 locations in Chicago, IL and Riverside, CA using measurements taken in December 2006 and April 2007. After adjustment for regional unspecified air pollutant concentration gradients, correlation of UFP with noise was Pearson $R = 0.31$ for Chicago and Pearson $R = 0.41$ for Riverside. Correlation of noise with UFP concentrations was higher within a 100-m buffer of the road (Chicago: Pearson $R = 0.37$; Riverside: Pearson $R = 0.58$) compared with outside the buffer (Chicago: Pearson $R = 0.08$; Riverside: Pearson $R = 0.50$).

### 3.4.4 PM Composition and Exposure Assessment

Compositional differences in ambient PM and ambient PM that has infiltrated indoors were discussed briefly in the 2009 PM ISA (U.S. EPA, 2009b). Several studies cited in the 2009 PM ISA found that $SO_4^{2-}$ comprised the largest proportion of ambient $PM_{2.5}$ exposure in studies from the eastern U.S., while a study in Denver found $NO_3^-$ to be the largest contributor to $PM_{2.5}$. Studies of differential infiltration of $PM_{2.5}$ by BC or OC found that BC contributed more to indoor $PM_{2.5}$ compared with OC. 2013–2015 composition data across the U.S. shows that, while there is still more $SO_4^{2-}$ in the east compared with the west, OC now is the most prevalent component of $PM_{2.5}$ in many areas across the country (Section 2.5.1.1.6).

This section provides new information on PM composition for $PM_{2.5}$, $PM_{10-2.5}$, and UFP from the peer-reviewed literature. Section 3.4.4.1 presents correlations between PM mass and composition from AQS and from the peer-reviewed literature. Section 3.4.4.2 is a new section of the ISA that presents data on studies of ROS exposure in the literature.

#### 3.4.4.1 Composition

Select epidemiologic studies of the health effects of PM exposure have examined potential associations between health effects and exposure to PM components (CHAPTER 5, CHAPTER 6,

CHAPTER 7, CHAPTER 8, CHAPTER 9, CHAPTER 10, and CHAPTER 11). These studies compare the effect estimates for exposure to PM components with health effect estimates for exposure to total PM, measured as ambient mass concentration (MC), NC, or personal exposure concentration. This section presents relationships between concentrations of total PM with PM components.

Figure 3-17 displays correlations for 24-hour ambient $PM_{2.5}$ mass concentration with mass concentration for select components of $PM_{2.5}$ measured from the AQS during 2013–2015 on an annual basis, and Figure 3-18 displays the correlations on a seasonal basis. Median correlations with $PM_{2.5}$ were ordered as OC > $SO_4^{2-}$ > EC > $NO_3^-$ > Cl > Si, with correlations above Pearson $R = 0.5$ for OC, $SO_4^{2-}$, EC, and $NO_3^-$. Sulfate, $NO_3^-$, and OC are most commonly a product of chemical reactions of air pollutants in the atmosphere, and PM produced during atmospheric chemistry is often in the fine size range (Section 2.2). The median correlation of $PM_{2.5}$ with Cl and Si was approximately Pearson $R = 0.2$. On a seasonal basis, correlations between $PM_{2.5}$ and $NO_3^-$ were lower during the spring and summer months, perhaps coinciding with less home heating fuel use during the summer. In the peer-reviewed literature (Figure 3-19), correlations of ambient $PM_{2.5}$ with ambient $SO_4^{2-}$ and $NO_3^-$, used as exposure concentration surrogates, were similarly high (Ito et al., 2011; Ostro et al., 2010; Ostro et al., 2009), but much greater variability in correlations were observed for ambient OC and more so for EC or BC (which were combined for presentation purposes). Median correlations were around 0.5 for most trace metals, but higher correlations were observed for S, Zn, and V in New York (Ito et al., 2011) and Southern California (Ostro et al., 2010; Polidori et al., 2009). The higher correlations for S are likely explained by $SO_4^{2-}$. Ito et al. (2011) and Polidori et al. (2009) attributed elevated correlations with Zn and V to residential oil combustion.

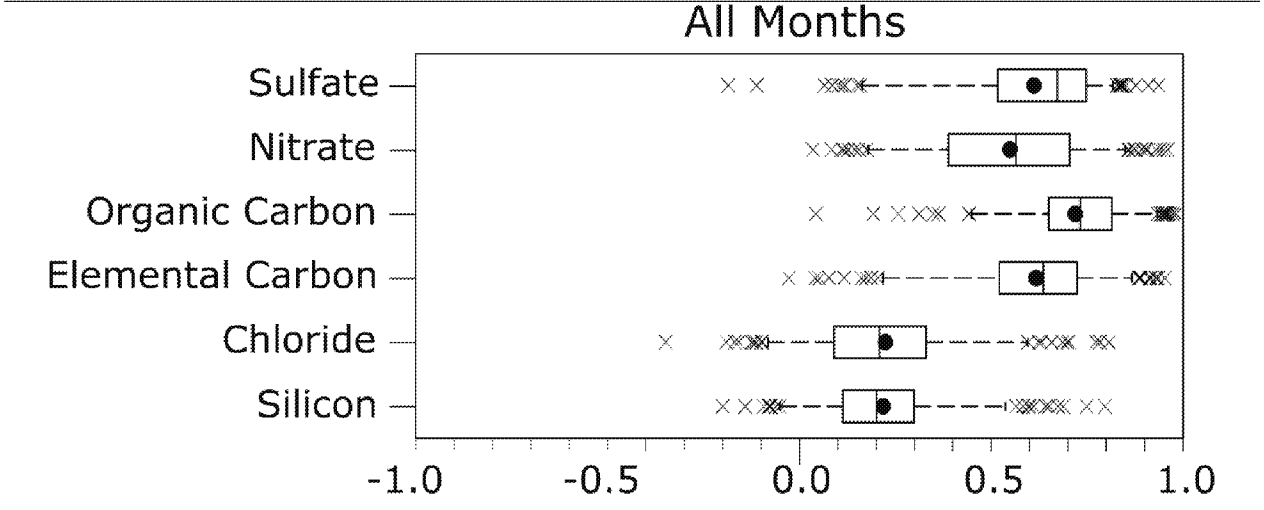



**Figure 3-17 Distribution of Pearson correlation coefficients for annual 24-hour average $PM_{2.5}$ mass concentration with mass concentration of $PM_{2.5}$ components from the Air Quality System during 2013–2015.**

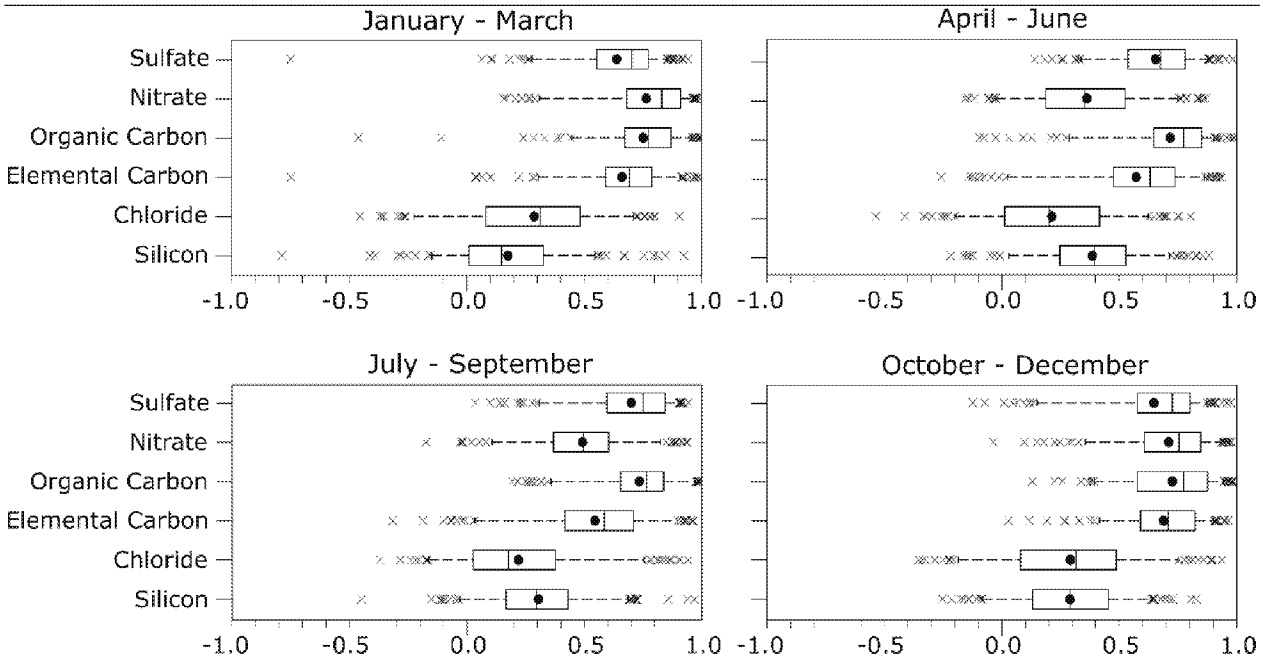
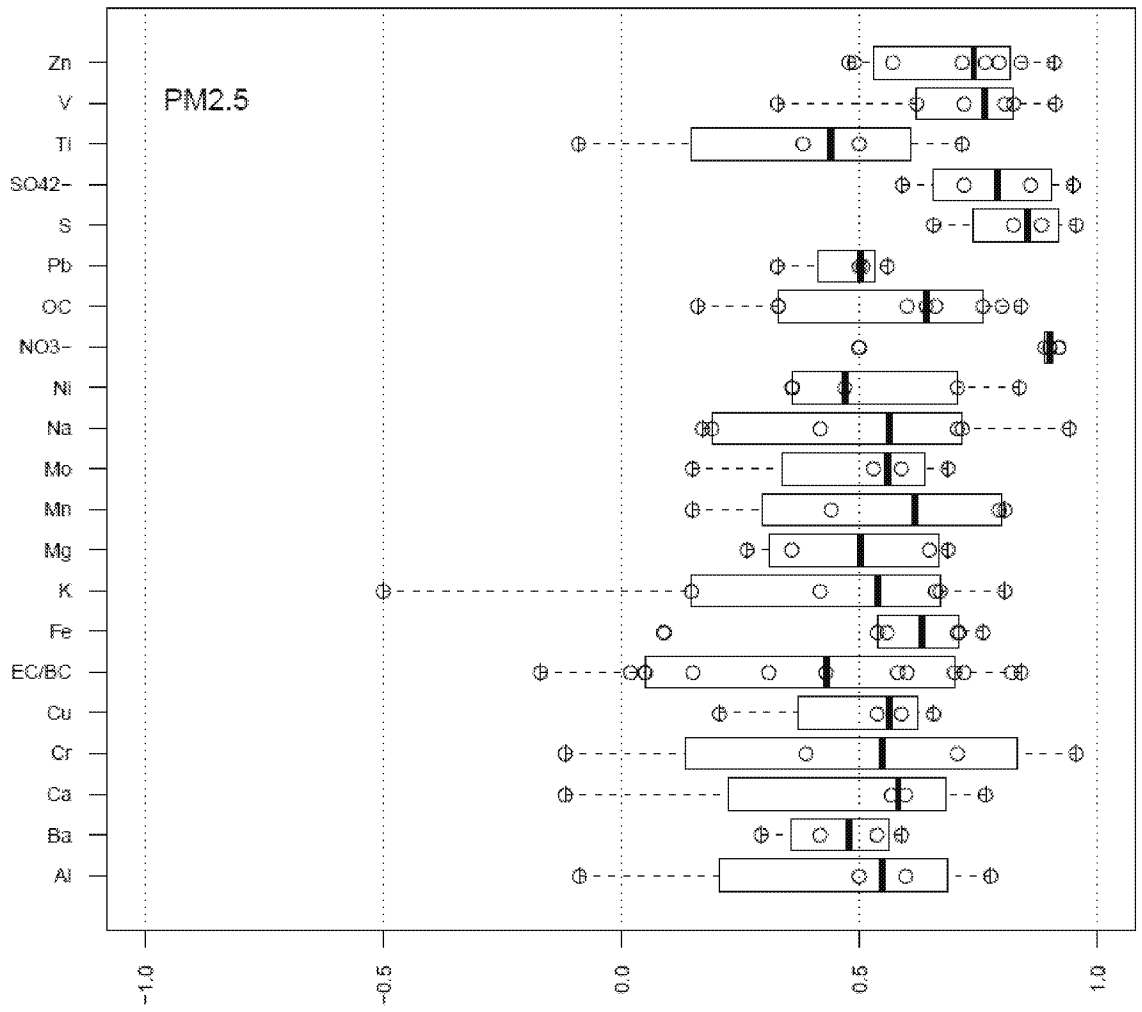



**Figure 3-18 Distribution of Pearson correlation coefficients for comparison of seasonal 24-hour average total $PM_{2.5}$ mass with mass concentration of $PM_{2.5}$ components from the Air Quality System during 2013–2015.**




Source: Permission pending, Polidori et al. (2009); Ito et al. (2011); Ostro et al. (2009); Raysoni et al. (2013); Zhang et al. (2016); Delfino et al. (2013); Delfino et al. (2010); Ostro et al. (2010).

**Figure 3-19 Distribution of Pearson correlation coefficients for annual 24-hour average total $PM_{2.5}$ mass concentration with mass concentration of $PM_{2.5}$ components from the peer-reviewed literature during 2013–2015.**

For $SO_4^{2-}$, OC, $NO_3^-$, and EC, site-specific correlations range from near Pearson $R = 1$ down to near Pearson $R = 0$ (Figure 3-17). This suggests spatial variability of the correlations between $PM_{2.5}$ and each component (Figure 3-20). Maps of Pearson correlations at AQS sites measuring $PM_{2.5}$ and components illustrate the level of variability for the four components. Correlations between $PM_{2.5}$ and $SO_4^{2-}$ are highest in the northeastern and Midwestern portions of the U.S. Correlations between $PM_{2.5}$ and $NO_3^-$ are highest in the West and markedly lower throughout the Southeast and Midwest. Correlations between $PM_{2.5}$ and EC appear highest in the West, possibly due to the influence of wildfire on $PM_{2.5}$ concentrations (Section 2.5.1.1.6).

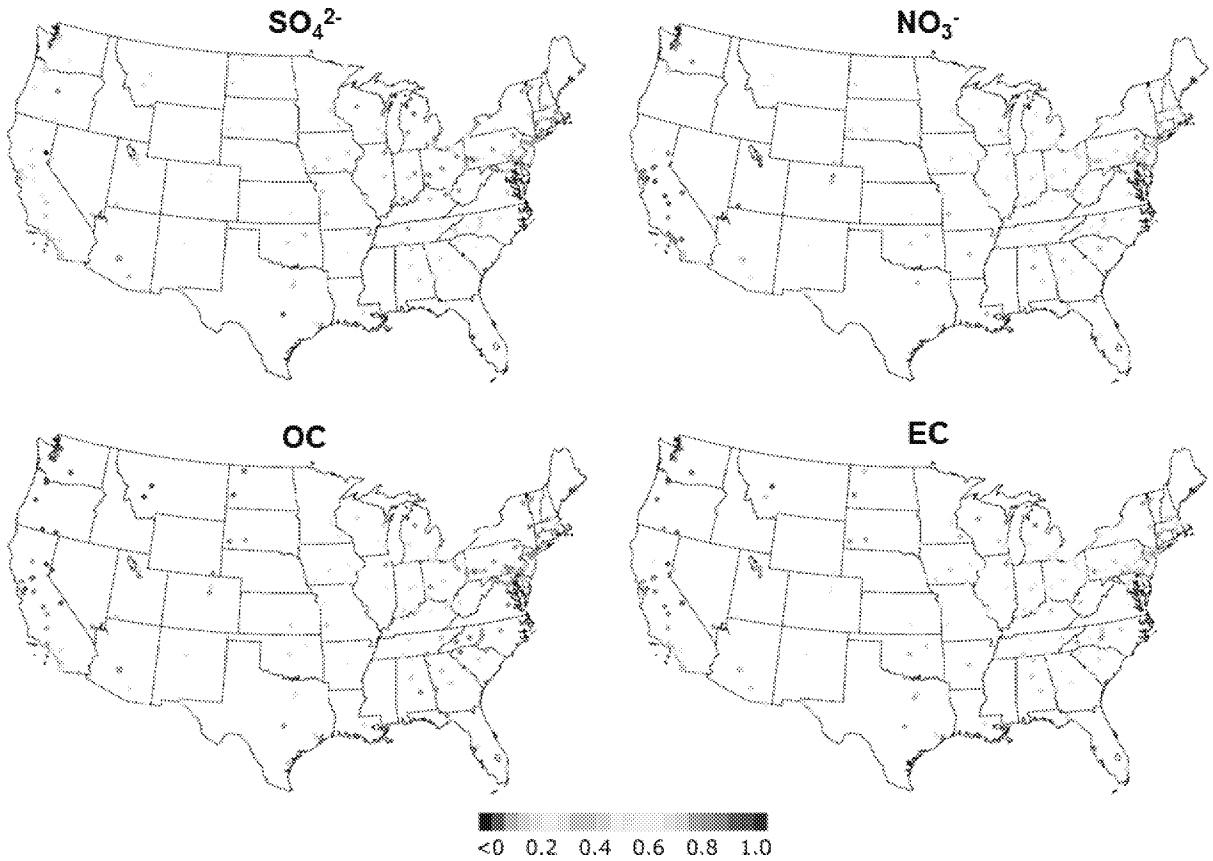



**Figure 3-20 Maps illustrating national-scale variability of Pearson correlation coefficients for comparison of seasonal 24-hour average total $PM_{2.5}$ mass concentration with mass concentration of $PM_{2.5}$ components from the Air Quality System during 2013–2015.**

Figure 3-21 displays annual correlations for 24-hour ambient $PM_{10-2.5}$ mass concentration with mass concentration for select components of $PM_{10-2.5}$ measured from the AQS during 2013–2015, and Figure 3-22 displays seasonal correlations. Median correlation of $PM_{10-2.5}$ mass concentration with Si was slightly lower than Pearson $R = 0.7$, while median correlations of $PM_{10-2.5}$ mass concentrations with the other $PM_{10-2.5}$ components were between Pearson $R = 0$ and Pearson $R = 0.3$. The difference between correlations for Si with those for the other components holds across seasons, with the highest correlation for Si and lowest correlations for all other components evident during the fall months (Figure 3-22). The higher correlation of $PM_{10-2.5}$ mass concentration and Si in $PM_{10-2.5}$ was likely due to the influence of dust, particularly in the Southwestern U.S. (Section 2.5). Figure 3-24 shows higher correlations in the Southwest, in support of this claim. Data for correlations between ambient $PM_{10-2.5}$ mass concentration and Si in $PM_{10-2.5}$ (for each of these studies, ambient $PM_{10-2.5}$ and components were measured by fixed-site monitors outside the location where personal samples were obtained, but no correlations were reported for personal samples) were not available in the literature for comparison (Raysoni et al., 2013; Delfino et al., 2010; Polidori et al., 2009), but median correlations for components reported were all less than Pearson $R = 0.5$ (Figure 3-23).

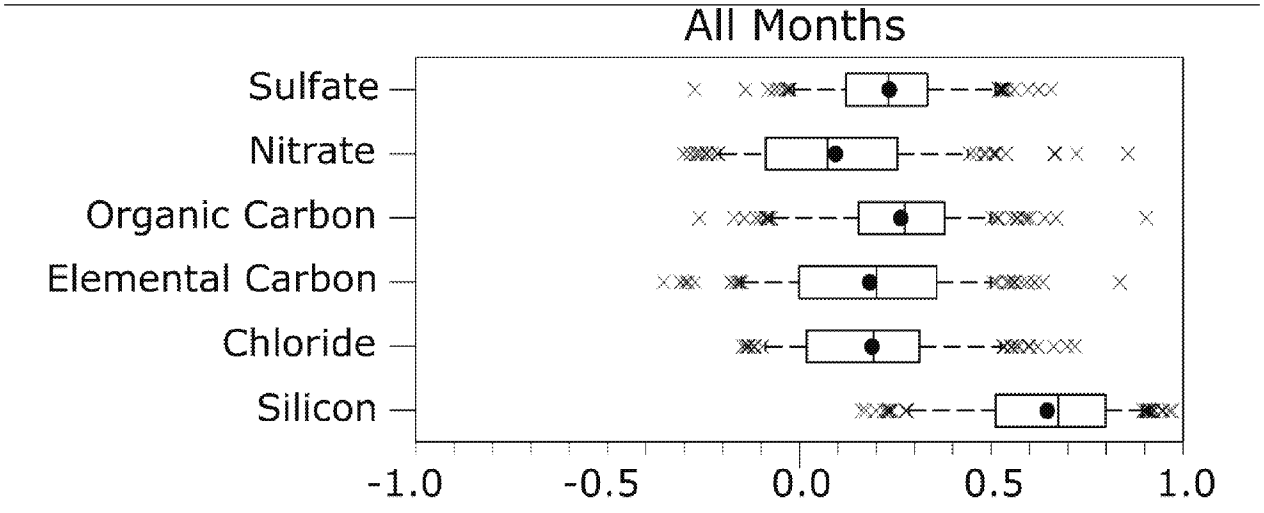



**Figure 3-21 Distribution of Pearson correlation coefficients for annual 24-hour average total mass concentration of $PM_{10-2.5}$ with mass concentration of $PM_{10-2.5}$ components from the Air Quality System during 2013–2015.**

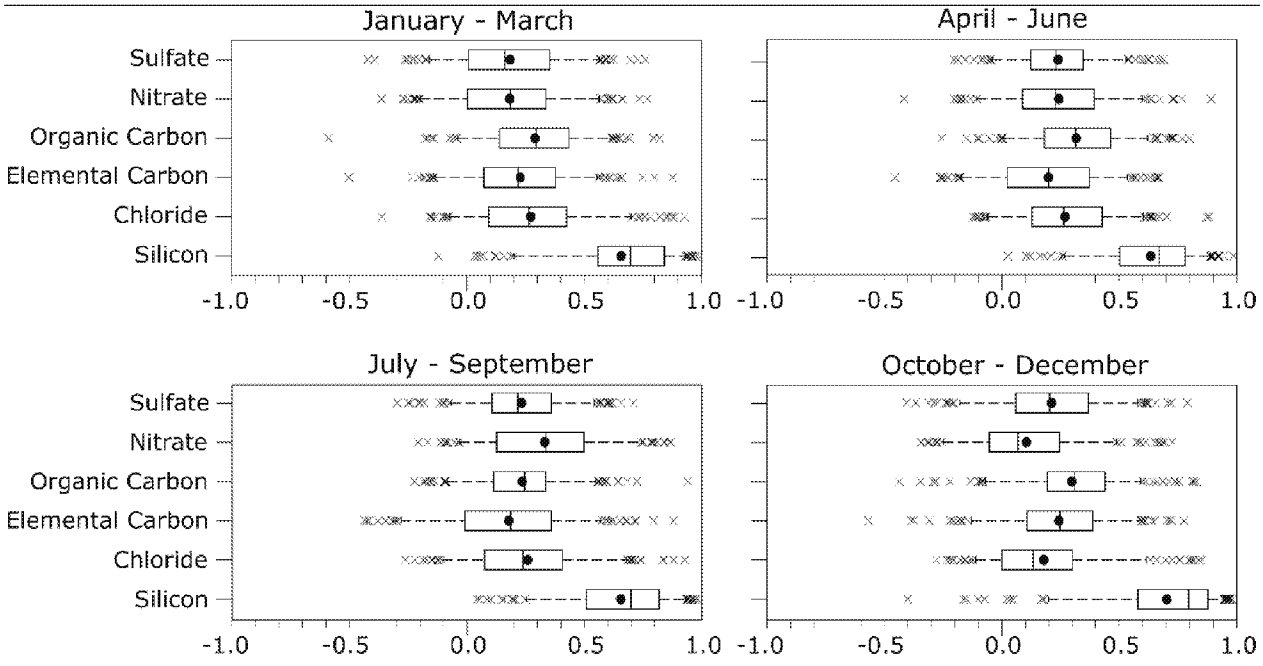
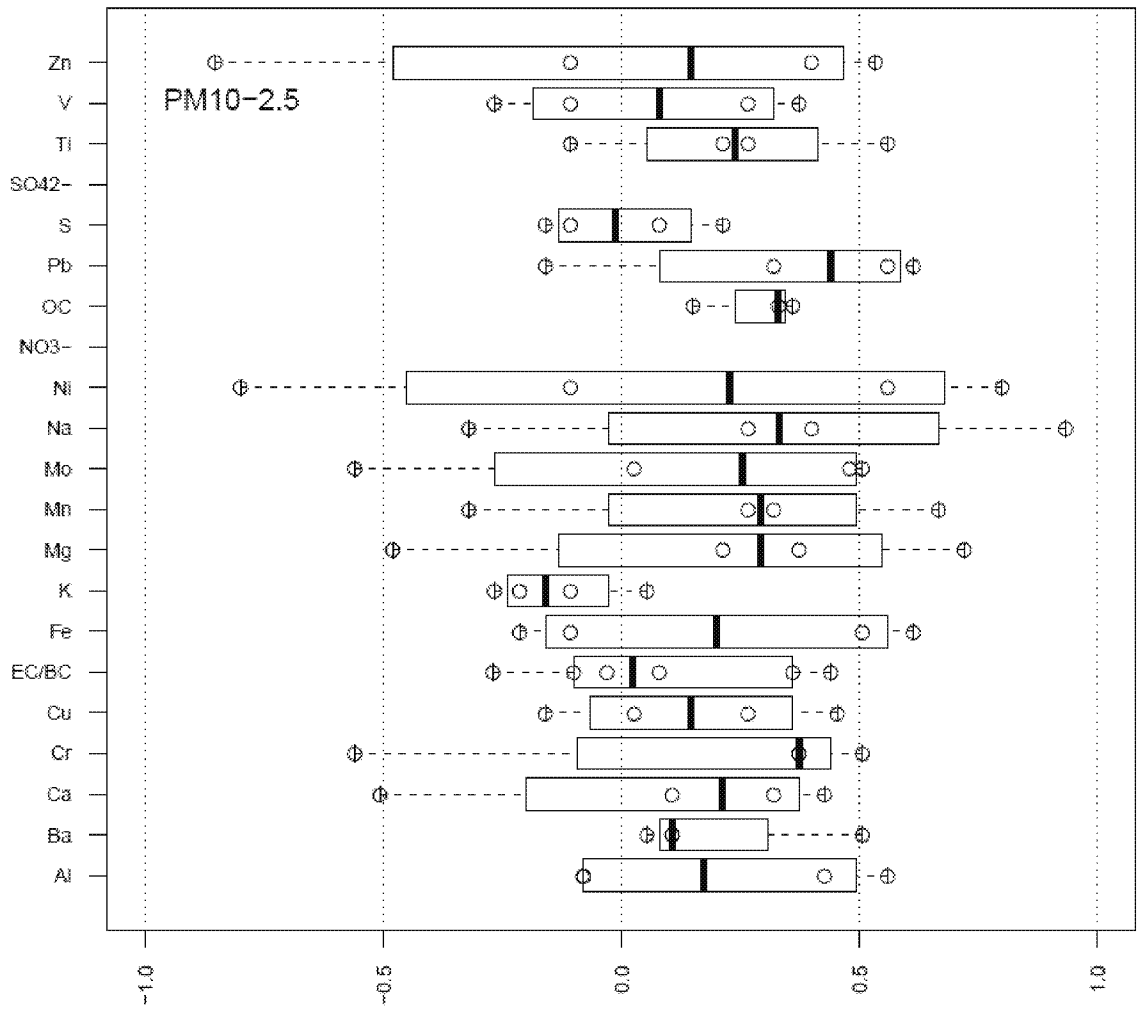



**Figure 3-22 Distribution of Pearson correlation coefficients for comparison of seasonal 24-hour average total $PM_{10-2.5}$ mass concentration with mass concentration of $PM_{10-2.5}$ components from the Air Quality System during 2013–2015.**




Source: Permission pending, Polidori et al. (2009); Raysoni et al. (2013); Delfino et al. (2010).

**Figure 3-23 Distribution of Pearson correlation coefficients for annual 24-hour average total $PM_{10-2.5}$ mass concentration with mass concentration of $PM_{10-2.5}$ components from the peer-reviewed literature.**

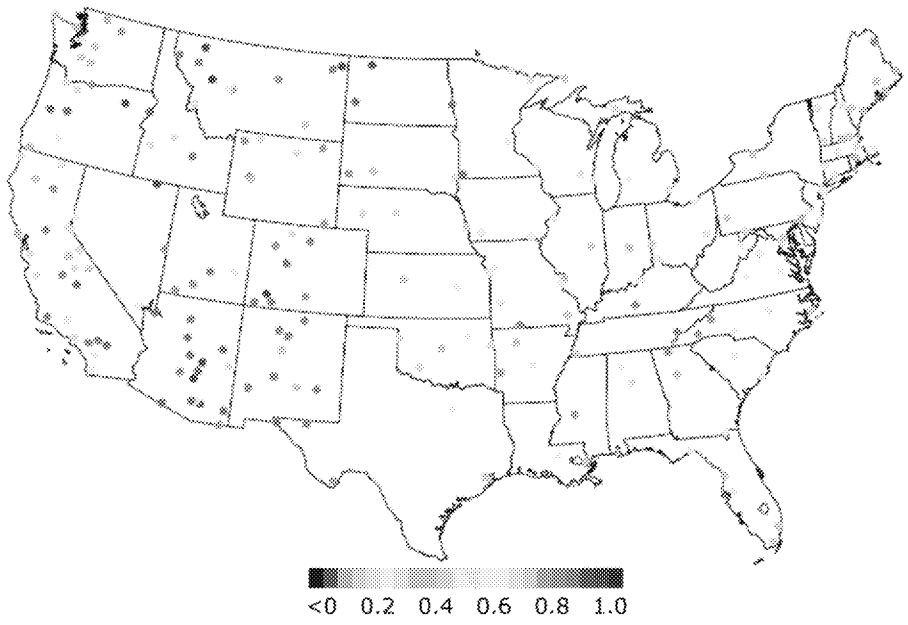
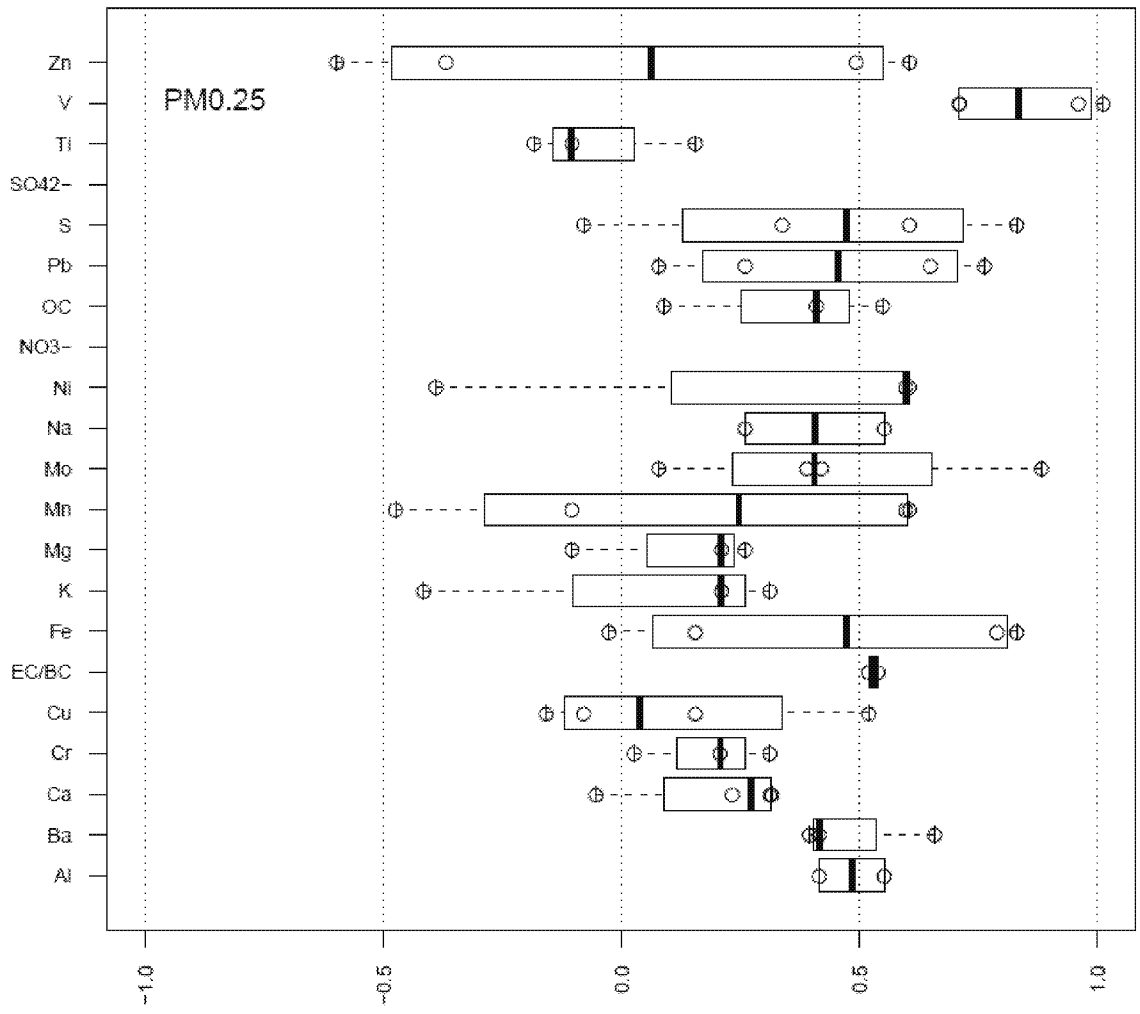



**Figure 3-24 Map illustrating national-scale variability of Pearson correlation coefficients for comparison of seasonal 24-hour average total $PM_{10-2.5}$ mass concentration with mass concentration of Si in $PM_{10-2.5}$ from the Air Quality System during 2013–2015.**

Exposure to UFP composition is informed by considering data for correlations of mass concentration for PM smaller than 250 nm ($PM_{0.25}$). These samples were measured using a cascade impactor, with concentrations of $PM_{0.25}$ components were calculated based on ambient fixed-site measurements for monitors placed outside retirement communities as surrogates for exposure concentration in Polidori et al. (2009) and Delfino et al. (2010), as shown in Figure 3-25. The highest median correlation was between $PM_{0.25}$ and V (Spearman $R = 0.8$), which tends to be present in heating oil and industrial waste (Polidori et al., 2009). Correlation between $PM_{0.25}$ and V was near Spearman $R = 1$ in the cool season and near Spearman $R = 0.7$ during the warm season, which is consistent with heating oil use. Medium correlations near Spearman $R = 0.5$ were reported for several components, including S (correlations with $SO_4^{2-}$ were not reported at the $PM_{0.25}$ size cut), Pb, OC, Ni, Na, Mo, Fe, EC/BC, Ba, and Al. Both studies took place in 2005–2007, and ultra-low sulfur diesel fuel was phased in between 2006 and 2010. Moderate correlations for $PM_{0.25}$ with S, EC/BC, OC, and Ba could be related to traffic (Polidori et al., 2009).




Source: Permission pending, Polidori et al. (2009); Delfino et al. (2010).

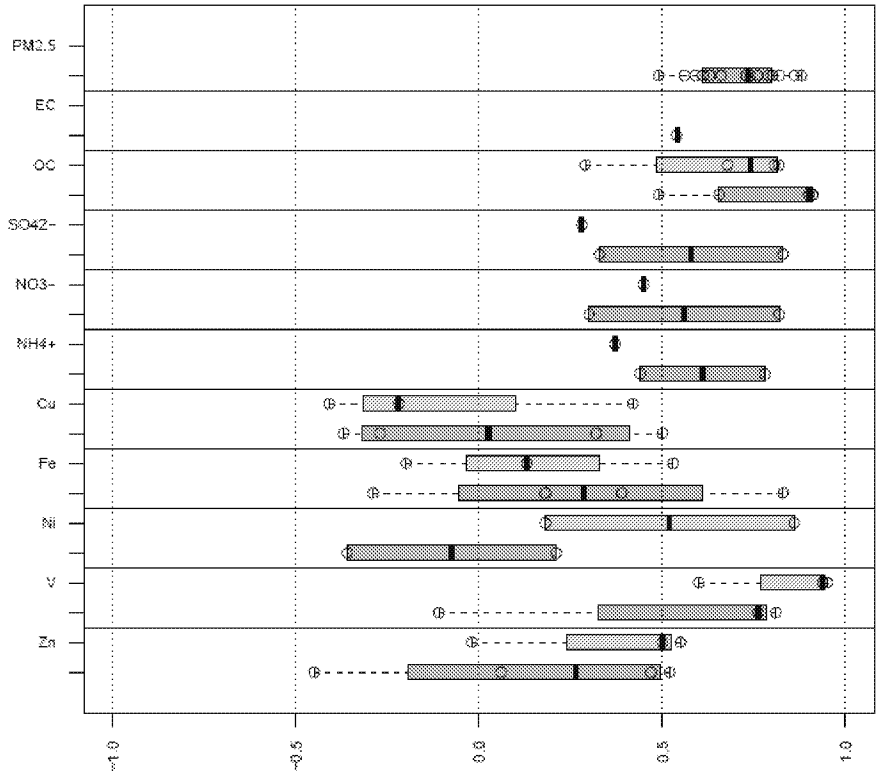
**Figure 3-25 Distribution of Pearson correlation coefficients for annual 24-hour average total $PM_{0.25}$ mass concentration with mass concentration of $PM_{0.25}$ components from the peer-reviewed literature.**

### 3.4.4.2 Reactive Oxygen Species

Recent exposure assessment studies inform biological plausibility discussions (Section 5.2.1, Section 5.3.1, Section 6.2.1, Section 6.3.1, and Section 10.2.1) because they measure oxidative potential

as a surrogate for oxidative stress. Oxidative stress and inflammation may be initiated by PM exposure, when a target site does not have enough antioxidant reserve to counteract the ROS. Oxidative stress can occur directly through redox reaction, or it can occur indirectly, where redox-inactive metals can form complexes with antioxidants so that the cell is then vulnerable to oxidation. The dithiothreitol (DTT) assay for measuring ROS inform PM's ability to cause oxidative stress directly [see Cho et al. (2005), Section 3.3.1.2]. Macrophage ROS assays [see Landreman et al. (2008), Section 3.3.1.2] provide a model of both direct and indirect oxidative stress, because both may occur in the model cell.

ROS activity for ambient PM is shown in Figure 3-26 through correlations of ROS macrophage and DTT assay results with mass concentration of $PM_{2.5}$, prevalent components (EC, OC, $SO_4^{2-}$, $NO_3^-$, and $NH_4^+$), and select trace metals (Cu, Fe, Ni, V, Zn) (Bates et al., 2015; Fang et al., 2015; Verma et al., 2009; Hu et al., 2008). Correlations between $PM_{2.5}$ mass concentration and DTT activity ranged from Pearson $R = 0.49$ to 0.88. No studies presented correlations between $PM_{2.5}$ mass and ROS activity based on the macrophage ROS assay, and limited data were available for the components presented. Most correlations were greater than 0.3 for EC, OC, $SO_4^{2-}$, $NO_3^-$, and $NH_4^+$. For trace metals, correlations ranged from positive to negative, where negative correlations imply that the ROS activity goes down with increasing concentration of the PM components or vice versa. In most cases, boxplots overlapped for the DTT and macrophage ROS assay, suggesting that both types of assay results covary similarly with measures of concentration for $PM_{2.5}$ components, despite the inability of DTT to capture indirect oxidation processes. These findings suggest that mass concentration of ambient $PM_{2.5}$ components may inform epidemiologic studies of oxidative stress and related effects. However, oxidative potential approaches are limited as a model of oxidative stress, because they do not reproduce the oxidative stress mechanisms. Moreover, macrophage ROS assay data are needed to correlate with ambient $PM_{2.5}$ mass concentration to consider if ambient $PM_{2.5}$ mass concentration is associated with direct and indirect ROS activity.




$PM_{2.5}$ = particulate matter with 50% aerodynamic diameter less than a nominal diameter of 2.5 μm; EC = elemental carbon; OC = organic carbon; $SO_4^{2-}$ = sulfate; $NO_3^-$ = nitrate; $NH_4^+$ = ammonium; Cu = copper; Fe = iron; Ni = nickel; V = vanadium; Zn = zinc.

Note: For each element, correlations obtained through the dithiothreitol assay are shown in orange at the bottom of each box and correlations obtained through the reactive oxygen species macrophage ROS assay are shown in light blue at the top of each box.

Source: Permission pending, Bates et al. (2015), Fang et al. (2015), Hu et al. (2008), Verma et al. (2009).

**Figure 3-26 Pearson correlations of ambient air measures of oxidative potential with $PM_{2.5}$ mass and $PM_{2.5}$ components.**

Personal exposure measurements were correlated to ROS activity for three studies of PM exposures in a school (Delfino et al., 2013) and retirement communities (Zhang et al., 2016; Delfino et al., 2010). In the school study, correlations ranged from Spearman $R = 0.77$ to 0.85 for the DTT assay's relationship to $PM_{2.5}$ mass, EC, OC, and water-soluble OC exposure concentrations. Similarly, correlations ranged from Spearman $R = 0.66$ to 0.86 for the same components for the macrophage ROS assay's relationship to $PM_{2.5}$ mass, EC, OC, and water-soluble OC exposure concentrations. The first retirement home study occurred between 2005 and 2007 and included Spearman correlations of macrophage ROS activity with $PM_{10-2.5}$, $PM_{2.5-0.25}$, and $PM_{0.25}$ mass exposure concentrations, along with

NC and components of EC, OC, BC, primary OC (POC), and secondary OC (SOC). Correlations of macrophage ROS activity with $PM_{10-2.5}$ and $PM_{2.5-0.25}$ were Spearman $R = 0.09$ and 0.07, respectively. Correlations of ROS activity with $PM_{0.25}$ mass exposure concentration (Spearman $R = 0.41$) and for NC (Spearman $R = 0.23$) were higher by comparison. EC, OC, BC, and POC had correlations of Spearman $R = 0.31$ to 0.40, while the correlation for SOC with ROS activity was 0.08.

Assays to measure ROS activity were recently evaluated for particles near the UFP size range. Zhang et al. (2016) correlated ROS activity of particulate matter smaller than 180 nm ($PM_{0.18}$) or of particulate matter between 180 and 250 nm ($PM_{0.25-0.18}$) with $PM_{2.5}$, BC, and components' exposure concentrations within the $PM_{0.18}$ and $PM_{0.25-0.18}$ size ranges. Correlation was Spearman $R = -0.17$ and 0.05, respectively for the DTT and macrophage ROS assays, for the correlation of $PM_{2.5}$ exposure concentration with ROS activity of $PM_{0.18}$. Correlation was Spearman $R = 0.20$ and 0.45 for the correlation of $PM_{2.5}$ exposure concentration with ROS activity of $PM_{0.25-0.18}$, so that ROS activity of $PM_{0.25-0.18}$ correlated more with $PM_{2.5}$ exposure concentration than did ROS activity of $PM_{0.18}$. Correlations among components of $PM_{0.18}$ exposure concentrations were higher for ROS activity of $PM_{0.18}$, but that pattern did not hold for ROS activity of $PM_{0.25-0.18}$. Additionally, larger differences were observed when correlations between exposure to mass concentration and ROS activity were measured by DTT (for DTT of $PM_{0.18}$, Spearman $R = 0.50$ to 0.86, and of $PM_{0.25-0.18}$, Spearman $R = 0.25$ to 0.62) than when they were measured by the macrophage ROS assay (for ROS of $PM_{0.18}$, Spearman $R = -0.02$ to 0.45, and of $PM_{0.25-0.18}$, Spearman $R = 0.09$ to 0.41). This may imply that for $PM_{0.25}$, mass exposure concentration of components may be associated with direct redox activity but not with indirect oxidation via antioxidant complexation. No correlations of $PM_{0.25-0.18}$ or $PM_{0.18}$ total mass exposure concentration were provided in the Zhang et al. (2016) study. However, the Delfino et al. (2010) study did provide correlation data for $PM_{0.25}$ and NC and found low-moderate correlations (Spearman $R = 0.41$ for $PM_{0.25}$ and Spearman $R = 0.23$ for NC), consistent with the correlations of the $PM_{0.18}$ and $PM_{0.25-0.18}$ components' mass exposure concentrations with the macrophage ROS assay results. Hence, multiple studies indicate that the macrophage ROS assay is a reliable indicator of oxidative potential.

### 3.4.5 Influence of Exposure Errors on Results from Epidemiologic Studies of Different Designs

Exposure measurement error, which refers to the biases and uncertainties associated with using concentration metrics as surrogates for the actual exposure of an individual or population (Section 3.2.1, Exposure Terminology), can be an important contributor to error in epidemiologic study results. Time-series studies assess the daily health status of a population of thousands or millions of people over the course of multiple years (i.e., thousands of days) across an urban area by estimating people's exposure using a short monitoring interval (hours to days). In these studies, the community-averaged concentration of an air pollutant measured at ambient monitors is typically used as a surrogate for individual or population ambient exposure. In addition, panel studies, which consist of a relatively small sample

(typically tens) of study participants followed over a period of days to months, have been used to examine the health effects associated with short-term exposure to ambient concentrations of air pollutants [e.g., Delfino et al. (1996)]. Panel studies may also apply a microenvironmental model to represent exposure to an air pollutant. A longitudinal cohort epidemiologic study, such as the American Cancer Society (ACS) cohort study, typically involves hundreds or thousands of subjects followed over several years or decades [e.g., Jerrett et al. (2009)]. Ambient concentrations are generally aggregated over time and by community as exposure surrogates.

Exposure error can bias epidemiologic associations between ambient pollutant concentrations and health outcomes and tends to widen confidence intervals around those estimates (Sheppard et al., 2005; Zeger et al., 2000). The importance of exposure error varies with study design and is dependent on the spatial and temporal aspects of the design. Other factors that could influence exposure estimates include topography of the natural and built environment, meteorology, instrument errors, use of ambient PM concentration as a surrogate for exposure to ambient PM, and the fact PM is one part of a complex mixture of pollutants. The following sections will consider various sources of error and how they affect the interpretation of results from epidemiologic studies of different designs.

### 3.4.5.1 Short-Term Exposure Studies

#### 3.4.5.1.1 Time-Series Studies

As discussed in the 2009 PM ISA (U.S. EPA, 2009b), in most short-term exposure epidemiologic studies, the health effect endpoint is modeled as a function of ambient exposure, $E_a$, which is defined as the product of $C_a$ and $\alpha$, a term encompassing time-weighted averaging of microenvironmental exposures and infiltration of PM (Section 3.2.2, conceptual model). Time-series epidemiologic studies capturing the exposures and health outcomes of a large cohort frequently use the ambient concentration at a fixed-site monitor or an average of ambient concentrations across monitors as a surrogate for $E_a$ in a statistical model (Strickland et al., 2011; Wilson et al., 2000). This is necessary due to the infeasibility of measuring personal exposures for studies involving thousands of participants. Moreover, for time-series epidemiology studies of short-term exposure, the temporal variability in concentration is of primary importance to relate to variability in the health effect estimate (Zeger et al., 2000). $C_a$ can be an acceptable surrogate if the ambient monitor captures the temporal variability of the true air pollutant exposure. Spatial variability in PM concentrations across the study area could attenuate an epidemiologic health effect estimate if the exposures are not correlated in time with $C_a$ when ambient monitoring is used to represent exposure in the statistical model. If exposure assessment methods that more accurately capture spatial variability in the concentration distribution over a study area are employed, then the confidence intervals around the health effect estimate may decrease.

In a time-series study of ED visits for cardiovascular disease, Goldman et al. (2011) simulated the effect of classical and Berkson errors due to spatiotemporal variability among ambient or outdoor (i.e., an ambient monitor situated outside the home) air pollutant concentrations over a large urban area. For 24-hour average $PM_{2.5}$, the relative risk (RR) per unit mass was negatively biased in the case of classical error (1.0094 compared to the base case of 1.0139) and negligibly positively biased in the case of Berkson error (1.0144). Negative bias means that the health effect estimate underestimates the true health effect. The 95% confidence interval range for RR per ppm of $PM_{2.5}$ was wider for Berkson error (0.0144) compared with classical error (0.0097). Similar results were obtained for $PM_{2.5}$ components ($SO_4^{2-}$, $NO_3^-$, $NH_4^+$, EC, and OC).

Recent studies have explored the effect of spatial exposure error on health effect estimates to test the appropriateness of using ambient monitoring for time-series studies. Goldman et al. (2010) simulated spatial exposure error based on a semivariogram function across monitor sites with and without temporal autocorrelation at 1- and 2-day lags to analyze the influence of spatiotemporal variability among ambient or outdoor concentrations over a large urban area on a time-series study of ED visits for cardiovascular disease. A random term was calculated through Monte Carlo simulations based on the data distribution from the semivariogram, which estimated the change in spatial variability in exposure with distance from the monitoring site. The average of the calculated random term was added to an ambient monitoring time series (considered in this study to be the base case) to estimate population exposure to $PM_{2.5}$ subject to spatial error. For the analysis with temporal autocorrelation considered, RR per ppm for 24-hour average $PM_{2.5}$ dropped slightly to 1.0126 (95% CI: 1.0113, 1.0139) when it was compared with the ambient monitor RR per ppm = 1.0139.[41] When temporal autocorrelation was not considered, RR per unit mass similarly dropped to 1.0123 for 24-hour average $PM_{2.5}$. The results of Goldman et al. (2010) suggest that spatial exposure error from use of ambient monitoring data results in biasing the health effect estimate towards the null to underestimate the true health effect, but the magnitude of the change in effect was small.

In another study analyzing the influence of spatiotemporal variability among ambient or outdoor concentrations over a large urban area on health effect estimates, Goldman et al. (2012) evaluated the effect of different types of spatial averaging on bias in the health effect risk ratio and the effect of correlation between measured and "true" ambient concentrations of $PM_{2.5}$ and $PM_{10}$ and other air pollutant measures. Concentrations were simulated at alternate monitoring locations using the geostatistical approach described above (Goldman et al., 2010) for the 20 county Atlanta metropolitan area for comparison with measurements obtained directly from monitors at those sites. Geostatistical-simulated concentrations were considered by the authors to be "true" in this study, and other exposure assessment methods were assumed to have some error. Five different exposure assessment approaches were tested: (1) using a single fixed-site ambient monitor, (2) averaging the simulated

[41] Note that 95% CIs were not reported for the ambient monitor RR or for the cases where temporal autocorrelation was not considered.

exposure concentrations across all monitoring sites, (3) performing a population-weighted average across all monitoring sites, (4) performing an area-weighted average across all monitoring sites, and (5) population-weighted averaging of the geostatistical simulation (see Table 3-10). Goldman et al. (2012) observed that the exposure error was somewhat correlated with both the measured and "true" values, reflecting both Berkson and classical error components. For the single fixed-site ambient monitor, the exposure errors had a moderate positive correlation with the measured value. For the other exposure concentration estimation methods, the exposure errors were moderately negatively correlated with the "true" value, while having positive but lower magnitude correlation with the measured value. Additionally, the exposure bias, given by the ratio of the exposure error to the measured value, was higher in magnitude at the single fixed-site monitor than for the spatial averaging techniques for $PM_{2.5}$. Hence, compared with other exposure assessment methods, the health effect estimate would likely have greater bias towards the null (i.e., underestimation of the true health effect estimate) with reduced precision when a single fixed-site monitor is used to measure $PM_{2.5}$ concentration as a surrogate for exposure. However, exposure error is likely to cause some bias and imprecision for other exposure surrogate methods as well.

**Table 3-10 The influence of exposure concentration metrics on error in health effect estimates.**

| Exposure Estimation Approach | Bias[(Z−Z*)/Z][a] | $R^2$(Z, Z*)[b] | *R*[(Z−Z*), Z*][c] | *R*[(Z−Z*), Z][c] |
|---|---|---|---|---|
| $PM_{2.5}$ | | | | |
| Fixed-site monitor | 0.21 | 0.76 | −0.10 | 0.41 |
| Unweighted average | 0.05 | 0.85 | −0.28 | 0.14 |
| Population-weighted average | 0.05 | 0.84 | −0.28 | 0.14 |
| Area-weighted average | 0.04 | 0.84 | −0.29 | 0.13 |
| Geostatistical model—population-weighted average | N/A | 0.87 | −0.38 | 0.00065 |

N/A = not applicable.

[a]Data provided by the authors for Figure 5 of Goldman et al. (2012).

[b]Data provided by the authors of Figure 4 of Goldman et al. (2012).

[c]Pearson correlation.

Note: Model errors were based on comparisons between measured data and simulated data at several monitoring sites. Errors were estimated for a single fixed-site ambient monitor, various monitor averages, and values computed from a geostatistical model. Z denotes the measured concentration, and Z* denotes the "true" concentration, considered here to be from the geostatistical model. Bias in the exposure concentration metric is given as the proportion of error between the measurement and true value to the measurement.

Source: Permission pending, Goldman et al. (2012).

In addition to the effect of the correlations and ratios themselves, spatial variation in their values across urban areas also impacts time-series epidemiologic results. The Goldman et al. (2010) and Goldman et al. (2012) findings suggest more Berkson error in the spatially resolved exposure concentration metrics compared with the fixed-site ambient monitor and more classical error for the fixed-site ambient monitor estimate compared with the other exposure assessment techniques. Hence, more bias would be anticipated for the health effect estimate calculated from the fixed-site ambient monitor, and more variability would be expected for the health effect estimate calculated with the more spatially resolved methods. Differences in the magnitude of exposure concentration estimates are not likely to cause substantial bias, but they tend more to widen confidence intervals and thus reduce the precision of the effect estimate (Zeger et al., 2000). The more spatially variable air pollutants studied in Goldman et al. (2012) also had more bias in the health effect estimates. This occurred across exposure assessment methods but was more pronounced for the fixed-site ambient monitoring data. Note that the Goldman et al. (2010), Goldman et al. (2011), and Goldman et al. (2012) studies were performed only in Atlanta, GA. These simulation studies are informative, but similar simulation studies in additional cities would aid generalization of these study results.

Dionisio et al. (2014) evaluated differences in $PM_{2.5}$ effect estimates derived from ambient monitors, an AERMOD air quality model to capture spatial variability, and a SHEDS personal exposure model incorporating infiltration and time-activity patterns for ZIP codes in Atlanta. They found that personal exposure model-based estimates were lower than ambient monitor and air quality model estimates, which were relatively similar to one another. The study also evaluated attenuation of health effect estimates in single-pollutant and copollutant models using a classical error attenuation factor relating the observed health effect estimate and health effect estimate that was designated by the authors to be "true". In single-pollutant models, using a fixed-site monitor reduced the size of the health effect estimate to about 80% of the effect estimate from the air quality model. The health effect estimate based on the fixed-site monitor was much more attenuated to approximately 25% of the health effect estimate when the personal exposure model was used for the exposure concentration estimate. The degree of attenuation was slightly greater in copollutant models with $SO_4^{2-}$ and $O_3$, and slightly less in a copollutant model with $NO_X$. Due to the more regional nature of PM, little spatial variability in the health effect estimates and degree of attenuation was observed. The findings of this study also suggest that PM is not as susceptible to spatially varying exposure error as locally-emitted pollutants such as CO and $NO_X$.

To account for temporal variability in exposure, Dominici et al. (2004) used spline functions to control for the temporal trend in exposure concentration and outcome in time-series studies. Szpiro et al. (2014) compared a version of this method with an approach to pre-adjust the exposure to account for the time trend, without need to account for the trend in the outcome, to reduce bias in the effect estimate. This method is particularly applicable for repeated-measure cohort studies, since it takes advantage of the additional exposure data available from more frequent pollutant measurements compared to the infrequent outcome and covariate measures.

Section 3.4.2.4 also describes the influence of instrument accuracy and precision on the relationship between ambient PM concentrations and personal exposure to ambient PM. Exposure measurement error related to instrument precision has a smaller effect on health effect estimates in time-series studies compared with error related to spatial gradients in the concentration because instrument precision would not be expected to modify the ability of the instruments to respond to changes in concentration over time. Goldman et al. (2010) investigated the influence of instrument error on health effect estimates in a time-series epidemiology study by studying differences in exposure concentration estimates and health effect estimates obtained using collocated monitors. In this study, a random error term based on observations from collocated monitors was added to an ambient monitor's time series to simulate population estimates for ambient air concentrations subject to instrument precision error in 1,000 Monte Carlo simulations. Virtually no change in the risk ratio was observed for 24-hour average $PM_{2.5}$; the RR per ppm with simulated instrument precision error was 1.0138 compared with RR per ppm = 1.0139 for the ambient monitor. The amount of bias in the health effect estimate related to instrument precision was very small.

As described in the 2009 PM ISA (U.S. EPA, 2009b), nonambient sources of PM include indoor combustion, cooking, cleaning, and other activities. However, such exposure is unlikely to be temporally correlated with ambient PM exposure (Wilson and Suh, 1997), and therefore would not affect epidemiologic associations between ambient PM and a health effect in a time-series study. In simulations of a nonreactive pollutant, Sheppard et al. (2005) concluded that nonambient exposure does not influence the health outcome effect estimate if ambient and nonambient concentrations are independent. Because personal exposure to ambient PM is some fraction of the ambient concentration, it should be noted that effect estimates calculated based on personal exposure rather than ambient concentration will be positively biased in proportion to the ratio of ambient concentration to ambient exposure, and daily fluctuations in this ratio can widen the confidence intervals in the ambient concentration effect estimate. Uncorrelated nonambient exposure will not bias the effect estimate but may also widen the confidence intervals (Sheppard et al., 2005; Wilson and Suh, 1997).

#### 3.4.5.1.2 Panel Studies

Panel or small-scale cohort studies involving dozens of individuals may use more individualized concentration measurements, such as personal exposures, residential fixed-site indoor or outdoor measurements, or concentration data from local study-specific monitors. Modeled concentrations are not typically used as exposure surrogates in panel epidemiologic studies. Probabilistic, distribution-based approaches are not designed to estimate exposures for specific individuals, such as might be needed for panel epidemiologic studies. Another main disadvantage of the modeling approach is that the results of modeling exposure assessment must be compared to an independent set of measured exposure levels (Klepeis, 1999). In addition, resource-intensive development of evaluated and representative model inputs

is required, such as human activity patterns, distributions of air exchange rate, and deposition rate. Therefore, modeled exposures have been used much less frequently in panel epidemiologic studies.

Panel studies using hourly or other subdaily measurements are used to evaluate subclinical health effects, such as biomarkers of inflammation [e.g., Dubowsky et al. (2006)]. Sensitivity to averaging time may be tested by fitting models with various averaging times to identify the time period most associated with effects. However, temporal variations in exposure and covariates (e.g., temperature, other pollutants) can lead to temporal variability in exposure measurement error. Malloy et al. (2010) proposed a wavelet approach to add time-varying data into the statistical model used in an epidemiologic study. Simulations adding exposure measurement error to an hourly $PM_{2.5}$ data set indicated that the fine-scale wavelets describing shorter-frequency variation captured most of the exposure error, with little error accounted for by the coarse wavelets. The standard moving average approach of fitting models with successively longer averaging times showed the greatest exposure error at shorter averaging times (less than 20−60 hours), while the effect of simulated error was similar across averaging times wavelet approach showed similar error over averaging times of 10 hours or greater. This suggests that the wavelet approach may be better able to identify associations with health effects over short averaging times (e.g., 24 hours or less).

To evaluate the effect of small-scale intraurban spatial variability on health effect estimates, Sarnat et al. (2012) considered the influence of local exposure concentration metrics on respiratory effect estimates for a panel of school children. This study was conducted along the U.S.-Mexico border in El Paso, TX and Ciudad Juarez, Mexico, and 48-hour average concentrations measured from fixed-site ambient monitors, monitors outside the children's schools, and monitors inside the children's schools were all used as surrogates for PM exposure concentration. For $PM_{2.5}$, slightly higher health effect estimates were observed for indoor monitors compared with outdoor and fixed-site ambient monitors (2.7, 2.3, and 2.4%, respectively), although confidence intervals overlapped. $PM_{10-2.5}$ had a higher health effect estimate for indoor than outdoor monitors (2.8 vs. 2.0%), again with overlapping confidence intervals. No fixed-site ambient $PM_{10-2.5}$ data were available. For both $PM_{2.5}$ and $PM_{10-2.5}$, multivariate models with both indoor and outdoor concentration only showed associations for indoor concentration. This effect was more pronounced for $PM_{10-2.5}$, which exhibits greater urban spatial variability than $PM_{2.5}$. The authors suggested that exposure measurement error could result in biasing the health effect estimate toward the null to underestimate the health effect, given the finding of higher health effect estimate for the outdoor $PM_{2.5}$ monitor compared with the outdoor $PM_{10-2.5}$ monitor.

### 3.4.5.2 Long-Term Exposure Cohort Studies

For cohort epidemiologic studies of long-term human exposure to PM, where the difference in the magnitude of the concentration is of most interest, if $C_a$ is used as a surrogate for $E_a$, then $\alpha$ can be considered to encompass the exposure measurement error related to uncertainties in the time-activity data and infiltration. Spatial variability in PM concentrations across the study area could lead to bias in the

health effect estimate if $C_a$ is not representative of $E_a$. This could occur if the study participants are clustered in a location where their PM exposure is higher or lower than the exposure estimated at a modeled or measurement site. There is limited information regarding whether $C_a$ is a biased exposure surrogate in the near-road environment for epidemiologic studies of long-term exposure.

Choice of exposure surrogate can influence error in the health effect estimate. For example, Baxter et al. (2010) calculated bias and RMSE for health effect estimates based on different exposure estimation methods including evaluated regression models, distance from a major road, and an indoor exposure model that accounts for factors such as seasonality in infiltration of ambient $PM_{2.5}$ and EC. The simulated indoor concentrations produced unbiased health effect estimates, while the other exposure surrogates typically (but not always) biased the health effect estimate towards the null to underestimate the true health effect and inflated the RMSE relative to that of the indoor model. Distance surrogates had much larger biases and RMSE compared with models containing $PM_{2.5}$ or EC concentration measures. Kioumourtzoglou et al. (2014) developed linear mixed effects models to calibrate exposure surrogates (fixed-site ambient monitor and monitor outside a residence) against what was considered by the authors to be "true" personal exposure to ambient $PM_{2.5}$, estimated by multiplying the fixed-site ambient $PM_{2.5}$ measurement by the ratio of personal to ambient $SO_4^{2-}$. The calibration coefficients indicated that the fixed-site ambient monitor only captured 31% of the "true" personal exposure to ambient $PM_{2.5}$, and the outdoor monitor captured 54% of the "true" personal exposure to ambient $PM_{2.5}$. Hence, in both cases, the exposure surrogate was lower than the sulfate-derived personal exposure.

Researchers have recently compared the choice of ground-based or satellite-based estimation methods on epidemiologic effect estimates. Jerrett et al. (2016) compared several residential exposure concentration estimation methods using ground-based data (i.e., monitor, meteorological, land use, or spatial information) or satellite data for a large subset of the ACS cohort (668,629 individuals). The authors found that although the various methods yielded similar median $PM_{2.5}$ exposure concentration estimates (approximately 12 $\mu g/m^3$), effect estimates for circulatory mortality during 1982–2004 were much lower for the satellite methods than the ground-based methods. Of the seven methods tested, the highest effect estimate was produced by a ground-data-only two-stage model consisting of LUR followed by a BME kriging model of the residuals; this method also had the best model fit. This model produced a relative risk (95% CI) of 1.14 (1.11–1.17) per 10 $\mu g/m^3$ $PM_{2.5}$, while the lowest relative risk was observed with one of the two satellite-only methods (RR = 1.02, 95% CI = 1.00–1.04). Jerrett et al. (2016) calculated the Akaike Information Criterion (AIC) to assess model fit and found a negative association between HR and AIC ($R^2 = 0.94$), which suggests that use of the satellite method alone produced an attenuated effect estimate. The LUR-BME method estimated exposure concentrations on a 30 × 30 m (0.03 × 0.03 km) grid, while this satellite-only method provided estimates on a 1 × 1 km grid. The results of the Jerrett et al. (2016) study suggest that exposure estimation methods incorporating locally available ground data may introduce less exposure error than remote sensing methods alone, but that satellite methods have the capability to identify associations when ground data are lacking.

Spatial resolution of the exposure concentration estimates has been evaluated to examine the influence of spatial exposure error in cohort studies. For example, Alexeeff et al. (2015) fit kriging and LUR models based on 100 or 500 monitoring sites [derived from a satellite downscaling approach described in Kloog et al. (2014) and Section 3.3.3] and estimated bias and uncertainty for each exposure concentration model used to compute health effect estimates for linear and logistic health effect simulations. For the LUR models, which had the highest model $R^2$ (71 to 84%) compared with the satellite-downscaling estimates, the effect estimates were biased away from the null to overestimate the health effect estimate in all cases. Bias in the linear models was reduced from 4−5% for LUR fit with 100 monitors to 1% for the LUR fit with 500 monitors, and confidence interval coverage increased from 48 to 68%. Bias in the logistic models was reduced from 3−4% for LUR fit with 100 monitors to 2% for LUR fit with 500 monitors, and confidence interval coverage increased from 91 to 94%. The kriging models had much lower model $R^2$ (24−44%). One kriging model fit to long-term average monitor data also produced bias away from the null to overestimate the health effect estimate that reduced with number of monitors, but with larger magnitude biases. The other produced bias mostly towards the null to underestimate the health effect estimate, with magnitude of bias increasing with increased number of monitors.

Gryparis et al. (2009) noted that smoothing of the true exposure concentration surface can cause Berkson error in the effect estimate. Gryparis et al. (2009) simulated three spatial surfaces of increasing variability and then tested five types of exposure concentration modeling approaches: plug-in exposure concentration estimation where the "true" exposure concentrations (as designated by the authors) are predicted by a smoothing model; plug-in exposure concentration estimation with variance correction; regression calibration using hold-out predictions, covariates, and observations; and two types of Bayesian surface models (full Bayesian and two-stage Bayesian approaches) fitting a joint model for the health and exposure concentration data. Simulation results produced negative biases to underestimate the health effect for the plug-in exposure concentration estimation methods with and without variance correction, and those biases became larger in magnitude with increasing spatial variability (for the plug-in method with variance correction, simulation results produced −57% bias for the smoothest surface and −419% bias in the most spatially variable surface). Likewise, the mean squared error (MSE) increased and confidence interval coverage decreased with increasing variability of the "true" exposure concentration surface. Biases and MSEs were much smaller in magnitude for the regression calibration and Bayesian exposure concentration assignment methods, and those biases were positive and so overestimated the health effect (maximum bias was 23% for the two-stage Bayes method for the most spatially variable exposure concentration surface). MSE for the regression calibration and Bayesian methods also increased with increasing variability of the "true" exposure concentration surface. Regression methods have also been applied to correct ambient monitor data or spatial modeling estimates of $PM_{2.5}$ exposure based on indoor $SO_4^{2-}$ to ambient $PM_{2.5}$ ratios in studies all-cause mortality (Hart et al., 2015a) and lung cancer (Hart et al., 2015b). In each study, the health effect estimate was lower when no exposure error correction method was applied. This implies that the smoother, non-corrected method introduced error into the exposure estimate that resulted in negative bias to underestimate the health effect.

The greater spatial characterization of $PM_{2.5}$ exposure concentration estimates from a combined satellite-LUR method with 50 m resolution developed by Kloog et al. (2011) resulted in higher mortality effect estimates compared with cohort studies using city-wide concentrations for the entire population based on a 10 km resolution grid (Kloog et al., 2013). This is consistent with a reanalysis of the ACS cohort conducted by Willis et al. (2003), which found that a subset analysis including only individuals living in a county with a sulfate monitor yielded an all-cause mortality effect estimate twice that for the entire cohort (1.5 vs. 1.25). The Kloog et al. (2013) study also found an effect of monitor distance, with a higher effect estimate for the population living within 20 km of a monitor than for those living farther away. This spatial influence on epidemiologic effect estimates is consistent with the null bias resulting from classical error.

The influence of spatial exposure error on health effect estimates varies with the study parameters, such as exposure model selection and location. Wu et al. (2011) compared health effect estimates for birth outcomes from four hospitals in Los Angeles and Orange Counties, CA given $PM_{2.5}$ concentrations as estimated using nearest monitors and the CALINE4 dispersion model. For preeclampsia, crude and adjusted odds ratios were consistently lower when the nearest monitor was used to estimate exposure concentration instead of the more spatially resolved dispersion model. Differences in the odds ratio for the two exposure concentration estimation methods were larger for Los Angeles County compared with Orange County. For Los Angeles County, the odds ratios were also below one when the nearest monitor was used, in contrast with Orange County, where the odds ratios were both above one. However, for preterm (<37 weeks gestation) and very preterm births (<30 weeks gestation), odds ratios were lower for the nearest monitor exposure concentration estimation method compared with the dispersion model in Los Angeles, but in Orange County, the opposite was observed. These findings indicate that higher spatial resolution may improve estimation of health effects.

Exposure error in studies of long-term exposure has the potential to be larger for $PM_{2.5}$ components than for $PM_{2.5}$ mass concentration, since the spatial variability of $PM_{2.5}$ components tends to be greater than for $PM_{2.5}$ mass concentration (Sun et al., 2013). Within components, the reported concentrations were also sensitive to the methods of measurement, with nearest monitor typically producing greater relative variability (measured as IQR/median) compared with IDW and city-wide average concentrations, respectively. Sun et al. (2013) compared statistical models of cardiovascular disease biomarkers associated with long-term exposure to $PM_{2.5}$ mass, EC, OC, Si, and S concentration using the nearest monitor, IDW, and city-wide average metrics. In general, effect estimates with city-wide averages tended to be lower in magnitude compared with the nearest monitor or IDW approaches for both the $PM_{2.5}$ mass and component metrics for one biomarker (CIMT) and for another biomarker (CAC) only for the Si component. Using finer-scale concentration estimates to approach the same problem, Kim et al. (2014) observed CIMT effects for Si but not EC. Little bias with $PM_{2.5}$ mass or S (as an indicator of $SO_4^{2-}$) concentration suggests that the less spatially variable metrics are less subject to bias related to exposure measurement error.

When a spatial concentration model, such as LUR or a spatiotemporal model, is used to develop a set of exposure concentration estimates for input into a long-term exposure epidemiologic study, minimizing error in the exposure or exposure concentration estimate does not always minimize error in the health effect estimate (i.e., β). Szpiro et al. (2011a) used simulation studies to evaluate the bias and uncertainty of the health effect estimate obtained when using correctly specified and misspecified exposure concentration models. The correct exposure concentration model was a spatiotemporal model with three geographic covariates while the misspecified model included only two of these three geographic covariates. In practice, covariates in spatiotemporal models may include variables such as population within a given buffer, proximity to industrial sources or highways, or building density. Szpiro et al. (2011a) did not explicitly state what the covariates were; as a statistical simulation study, the objective was to explore the impact of removing from the model a geographic covariate that may influence the exposure concentration. They estimated the exposure concentration model parameters using monitor data and predicted exposure concentrations at subject locations. They studied two conditions: where the variation in the third covariate was identical in the monitor and subject data versus where it was much smaller in the monitor data than in the subject data. Szpiro et al. (2011a) showed that prediction accuracy of the exposure concentration estimate was always higher for the correctly specified model compared with the misspecified model. The health effect estimate had better properties (lower RMSE) for the correct model when the third covariate had identical variability in the monitor and subject data. However, when the third covariate was much less variable in the monitor data, then the health effect estimate had better properties for the misspecified model. The results of Szpiro et al. (2011a) demonstrate one situation where use of a more accurately defined exposure concentration metric does not improve the health effect estimate.

Another simulation study evaluating the influence of exposure estimation methods on bias in health effect estimates considered the joint effect of exposure measurement error and confounding (Cefalu and Dominici, 2014). Exposure measurement error due to spatial variability in ambient concentrations or land use variables is often accounted for by exposure prediction models, such as LUR. Health effect models then may adjust for some of these same covariates as a means of reducing confounding of the effect estimate. Cefalu and Dominici (2014) demonstrated that if covariates are included in the exposure prediction model, but not the health effect model, the magnitude of bias in the health effect estimate is always increased relative to the simulated "true" exposure (as designated by the authors). The bias may be in either direction, depending on which covariates are omitted. To eliminate this bias, all potential confounders included in the health model must be included in the exposure prediction model, unless they are uncorrelated with exposure. Their simulation compared models with increasing numbers of covariates, and they found that in some situations the bias increased despite an increase in $R^2$, a similar result to the Szpiro et al. (2011a) study in which an improved exposure concentration metric did not improve the health effect estimate. One difficulty in applying these results to interpret epidemiologic study results is the uncertainty regarding the proper set of confounders to be included in the exposure and health models. While the Szpiro et al. (2011a) and Cefalu and Dominici (2014) simulations were for a generic air pollutant, they are relevant to spatially variable $PM_{10-2.5}$ or UFP.

Preferential sampling may occur when the exposure concentration model is fit to a set of spatial data, and exposures at other locations in the domain are not well represented. Sheppard et al. (2012) performed a series of simulations to study successively greater spatial correlations between monitors and study participants using kriging and nearest monitor to estimate $PM_{2.5}$ exposure concentration. Bias between the health effect estimate of the "true" exposure concentration (as designated by the authors) was compared with that derived from the kriged or nearest monitor exposure concentration estimates. Sheppard et al. (2012) found that bias decreased as spatial correlation between the "true" exposure concentration and the modeled exposure concentration increased. Both the kriging and nearest monitor exposure concentration models caused the coverage of the 95% confidence interval to be underestimated, but the underestimation was greater for nearest monitor. Furthermore, underestimation of the confidence interval became smaller with increasing spatial dependence of the "true" and modeled exposure concentrations. These results suggest that correlation between the "true" and modeled exposure reduces bias in the health effect estimate and reduces underestimation of variability in the health effect estimate. Lee et al. (2015) simulated several scenarios in which spatial variability explained successively larger portions of the exposure concentration variability to test for the effect of preferential sampling. Lee et al. (2015) also compared geospatial models of $PM_{2.5}$ components EC and S fit with the national network (urban and rural), CSN (urban), and IMPROVE (rural) networks and found large differences in the modeled exposure concentration surface. These results support the point that the nature of the monitors is important in deriving the surface. In general, Lee et al. (2015) found that the more preferential sampling occurred, the larger the relative bias and standard error of the effect estimate. In practice, studies of LUR have shown that fitting a model in one city and then applying it to another city can lead to large errors (U.S. EPA, 2016). The results of Lee et al. (2015) would imply that this practice would add error to the effect estimate.

Error correction is a relatively new approach to estimate the correct the classical-like standard error of exposure estimates and potentially to correct for bias in the exposure estimates used in statistical models for longitudinal cohort studies (Szpiro et al., 2011b). Szpiro and Paciorek (2013) and Bergen and Szpiro (2015) established that two conditions must hold for the health effect estimate to be predicted correctly: the exposure concentration estimates from monitors must come from the same underlying distribution as the true exposure concentrations, and the health effect model adjusts for confounding in the population. Szpiro and Paciorek (2013) performed several simulations to investigate what happens when these conditions are violated. In one set of simulations, the distribution of the exposure concentration was varied. When the assigned exposure concentration measurements were set to be uniform across space, the health effect estimate was biased away from the null (i.e., overestimated the health effect) with different standard error compared with the case when the exposure subjects were collocated with the study participants. When the model was misspecified, the health effect estimate was biased towards the null (i.e., underestimated the health effect) with different standard errors compared with the correctly specified model. Bias correction and bootstrap calculation of the standard errors improved the model prediction, even when the "true" model (as designated by the authors) contained several degrees of freedom. Spiegelman (2013) noted that the new measurement error correction methods developed by Szpiro and

Paciorek (2013) are a version of regression calibration. Bergen et al. (2013) applied error correction to models of long-term exposure to $PM_{2.5}$ components (EC, OC, Si, and S). They found that exposure errors in the EC and OC models were almost pure Berkson errors, so that the bootstrap calculation of the standard errors did not improve the estimates. Si and S were influenced by Berkson-like error, and bootstrap simulation of the standard errors was used for error correction. Absence of notable bias supports the observation of negligible classical-like error in the Si and S exposure concentration estimates.

In the case of long-term exposure cohort studies, nonambient contributions to the total personal exposure measurements would be expected to widen the confidence interval around the health effect estimates by adding noise to the exposure signal. Also, addition of any non-negative nonambient component to the personal exposure measurement would result in an underestimate of exposure to ambient PM, because the average total personal PM exposure would have to be either equal to or greater than the average personal exposure to ambient PM. This exposure error could bias the health effect estimate towards the null to underestimate the true health effect.

## 3.5 Summary

The exposure assessment chapter in the 2009 PM ISA (U.S. EPA, 2009b) synthesized a plethora of new research on PM, most of which focused on $PM_{2.5}$. The exposure assessment chapter in the 2009 PM ISA found that $PM_{10-2.5}$ tended to be more spatially variable than $PM_{2.5}$ at microscale, neighborhood scale, and urban scale, because $PM_{10-2.5}$ was more sensitive to local sources and loss processes, such as gravitational setting. UFP was also noted to be more spatially variable due to growth processes, but fewer data were available. Secondary production of $PM_{2.5}$ was noted to contribute to the relatively lower heterogeneity in its spatial concentration distribution. Similarly, infiltration was found to vary with particle size fraction, with the greatest infiltration factors occurring for $PM_{2.5}$ and infiltration decreasing with increasing particle size, due to surface impaction of $PM_{10-2.5}$ during the infiltration process. Source apportionment studies for $SO_4^{2-}$, as a marker of ambient $PM_{2.5}$, were presented as a method for distinguishing personal exposure to ambient $PM_{2.5}$ from total $PM_{2.5}$ exposure. Other components, such as EC and OC, were found not useful for apportionment of ambient $PM_{2.5}$ exposure, given their indoor sources. Spatial variability in PM concentration was noted to add uncertainty to exposure estimates.

Errors and uncertainties in the exposure assessment methods can add bias and uncertainty to health effect estimates from epidemiologic studies on the health effects of PM exposure. With regard to use of exposure surrogates in epidemiologic studies, the 2009 PM ISA (U.S. EPA, 2009b) noted that separating total PM exposure into ambient and nonambient components reduces uncertainty in health effects estimates. The 2009 PM ISA also noted that time-series studies of short-term $PM_{2.5}$ exposure generally use concentration data from fixed-site monitors as surrogates for exposure concentration, based on the assumption that temporal variability is captured at the monitor. Panel studies utilizing personal $PM_{2.5}$ exposure measurements found associations between short-term ambient $PM_{2.5}$ exposure and health

effects, and those findings were strengthened by focusing on the ambient component of exposure. It was noted that long-term $PM_{2.5}$ exposure studies produced health effects estimates that were most accurate when the PM concentration distribution does not vary substantially in space. Findings from the recent literature build from these results.

Fixed-site monitoring is still frequently utilized for exposure concentration surrogates for $PM_{2.5}$ (Section 3.3.1.1). Fixed-site monitoring data for $PM_{10-2.5}$ must be used with more caution. Generally, dichotomous samplers produce the most reliable measurements of $PM_{10-2.5}$ for use in exposure studies. Collocated $PM_{10}$ and $PM_{2.5}$ monitors used to calculate $PM_{10-2.5}$ concentration by difference can have higher errors and uncertainties due to differences in flow rates for the two instruments, while differences between $PM_{10}$ and $PM_{2.5}$ taken over a county or city to estimate $PM_{10-2.5}$ concentration has higher errors and uncertainties. CPCs are most commonly used to measure UFP. Some portion of the UFP size distribution may be omitted when using CPCs, since they do not typically measure particles smaller than 10 nm.

Substantial advances to exposure modeling have been made in recent years (Section 3.3.2). Spatial interpolation methods, LUR, dispersion models, and CTMs were already commonly used to estimate $PM_{2.5}$ exposure concentration. Improvements in modeling the OC component of $PM_{2.5}$ have improved the accuracy of CTMs in recent years. Additionally, hybrid approaches drawing input from CTMs, satellite observations of AOD, surface measurements of PM concentration, and land use variables data have been combined into spatiotemporal models. Microenvironmental exposure models have also been applied with input concentrations from these methods for comparison in epidemiology studies. The majority of studies using these methods are applied to model $PM_{2.5}$. These methods are employed less frequently to estimate $PM_{10-2.5}$ and UFP exposure concentration, related in part to less availability of input data. Epidemiologic study design influences selection of exposure concentration estimation methods.

Copollutant confounding of the PM health effect estimate may occur if exposure to the copollutants and their relationships to the health effect of interest are both correlated with PM exposure (Section 3.4.3). Median correlations of 24-hour ambient $PM_{2.5}$ with concentrations of ambient CO, $NO_2$, and $O_3$ during 2013–2015 were as high as Pearson $R = 0.5$, and upper correlations reached near 1. Copollutant correlation varied with season (highest for $O_3$ in summer and for CO and $NO_2$ in winter). Median correlations of 24-hour ambient $PM_{10-2.5}$ concentrations during the same time period were as high as Pearson $R = 0.4$, and upper correlations typically below Pearson $R = 0.7–0.8$. Median correlations between $PM_{2.5}$ and $PM_{10-2.5}$ range between 0.2 and 0.5, with higher values in summer and fall. Correlation data for UFP were very limited, but they indicate correlations as high as Pearson $R = 0.5$ for $NO_2$ and $NO_X$, which are also traffic-related pollutants. Moderate-to-strong correlations may introduce a greater degree of confounding into epidemiologic study results, depending on the relationship between the copollutants and the health effect of interest.

Ambient PM data from fixed-site monitors continue to be commonly used in health studies as a surrogate for PM exposure concentration (Section 3.3.1.1). Advantages to using fixed-site monitoring

data are that they provide a long-term record of concentration trends and they undergo rigorous quality assurance if FRMs or FEMs are used. The concentration profile of $PM_{2.5}$ tends to be less variable across the urban or neighborhood scale compared with $PM_{10-2.5}$ or UFP. Therefore, ambient $PM_{2.5}$ concentrations estimated at fixed-site monitors often provide a reasonable representation of exposure concentrations throughout the study area (Section 3.4.2.2). However, the higher degree of spatial variability in ambient $PM_{10-2.5}$ and UFP across an urban area may not be captured by a fixed-site monitor. Uncharacterized variability in a time-series of exposure concentrations across space, resulting from use of fixed-site monitoring data, in a time-series study of $PM_{10-2.5}$ or UFP exposure may attenuate health effect estimates, so that the health effect estimate underestimates the true health effect (Section 3.4.5.1). Bias may occur in either direction for long-term exposure studies, depending on whether the fixed-site monitor is over- or underestimating ambient $PM_{10-2.5}$ or UFP exposure concentration for the population of interest (Section 3.4.5.2). In all study types, use of fixed-site monitoring ambient $PM_{10-2.5}$ or UFP concentrations in lieu of the true exposure is expected to widen confidence intervals beyond what would be obtained if the true exposure were used. Personal monitors directly measure PM exposure, but they produce a relatively limited data set, making them most suitable for panel epidemiologic studies (Section 3.4.5.1.2). Without accompanying time-activity data, ambient PM exposure cannot be distinguished from personal PM exposure in personal monitoring studies (Section 3.4.2.1).

When spatial variability of exposure concentration surfaces is not accurately modeled, the health effect estimate tends to be biased towards the null with decreased probability that the confidence intervals contain the true health effect. Bias towards the null means that the health effect estimate is underestimating the true health effect. This is particularly true when the actual spatial variability is much higher than what is represented by the model (Section 3.4.5.2). Hybrid models typically have good cross-validation, especially for $PM_{2.5}$, and have the potential to reduce exposure measurement error and resulting bias and uncertainty in health effect estimates produced by epidemiologic models of long-term exposure to PM, even for spatially-varying size fractions and components. Bias correction and bootstrap calculation of standard errors have also been shown to improve health effect estimate prediction from spatiotemporal models when the exposure estimates have a classical-like error structure. When the exposure estimates have a Berkson-like error structure, health effect estimates would only be expected to improve when model covariates are chosen so that the statistical distribution of the modeled exposure concentrations is close to the distribution of the true exposure concentrations.

In summary, exposure error tends to produce underestimation of health effects in epidemiologic studies of PM exposure, although bias in either direction can occur. New developments in PM exposure assessment, including hybrid spatiotemporal models that incorporate satellite observations of AOD, land use variables, surface monitoring data from FRMs, and/or CTMs, have led to improvements in spatial resolution of the $PM_{2.5}$ concentration surface. These advancements have reduced bias and uncertainty in health effects estimates. However, high correlations with some gaseous copollutants necessitate evaluation of the impact of confounding on health effects estimates, using two-pollutant models to ascertain robustness of epidemiologic study results. $PM_{10-2.5}$ and UFP concentrations are typically more

spatially variable than $PM_{2.5}$ concentrations, and concentration data for those size fractions are less frequently available as model input or for use in validating hybrid models. As a result, there is typically less uncertainty in health effect estimates derived from both monitored and modeled exposure estimates for $PM_{2.5}$ compared with $PM_{10-2.5}$ and UFP.

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